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Stick-slip response in electrorheological fluids

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Experimental observations of stick-slip friction are reported in an electrorheological fluid. The results are consistent with shear-induced phase transitions between solid and liquid phases. During the slip, there is a liquid flowing between two columnar solid phases. When the liquid turns solid (reconnecting the columns), the structure supports a shear, and thus sticks.

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I. INTRODUCTION

Electrorheological (ER) response is a change in the rheology of a material in the presence of an electric field. ER fluids, discovered by Winslow [1], are suspensions of polarizable particles in dielectric liquids. The choices of particles and base fluid, as well as the magnitude of the electric field, dramatically affect the rheological properties of these materials. Recent studies of ER fluids provide a reliable model of their behavior [2–6].

Upon application of an electric field, the particles develop an electrostatic polarization due to the dielectric mismatch between the particles and the fluid. This polarization is further enhanced by the presence of other particles, since they intensify the local electric field experienced by any one particle. The forces due to polarization interactions cause the formation of particle chains aligned with the electric field. These chains then interact; the net result of which is the formation of columns. Thus, the suspension of randomly distributed particles develops a microstructure when in an electric field. The microstructure of a suspension is intimately related to its bulk rheological properties, so the formation and strength of these columns strongly affect the viscosity of an ER fluid. In common ER fluids of today, the effective viscosity of the suspension can be varied by orders of magnitude by varying the applied electric field. This response is both rapid and reversible.

In the last few years, Tao and his collaborators have described the nature of ER fluids in terms of electric-field-induced solidification [7,8,2,3]. There is a critical value of the electric field E_c above which the ER fluid turns into a solid. Physically, this solid can be characterized by a yield stress that rises as the electric field increases above E_c. The structure of the suspension microstructure in the solid phase has been pursued by numerous researchers. Halsey and Toor [4] assert that the thermodynamic ground state of an ER suspension in a high electric field is determined by a phase separation into low-density and high-density phases. In a parallel plate geometry this is manifested in the formation of columns of particles. Several computer simulations have verified this columnar structure [5,6]. Recently, Tao, and Sun [3] predicted a body-centered tetragonal (bct) structure in these columns and Chen, Zitter, and Tao [9] presented experimental evidence of this structure using laser diffraction.

The time scales for the formation of particle microstructure have been discussed theoretically by Halsey and Toor [10] and dynamic computer simulations of these times have been carried out by Klingenberg, van Swol, and Zukoski [5] and Bonnecaze and Brady [6]. A crude calculation of the time to form columnar structures can be made based on the assumption that Brownian diffusion drives the motion of the particles and the polarization forces only "steer" the particles. The rms distance that a particle travels is governed by the diffusion equation $\overline{\Delta x}=2Dt$, where D is the diffusion coefficient. D depends on the viscosity, η and the particle radius a; $D=kT/(6\pi\eta a)$. This predicts the relaxation time $\tau\sim 3\pi\eta a l^2/kT$, where l is the mean distance between particles. For typical ER materials, this time is on the order of 20 ms. This is in reasonable agreement with the computer simulations and experimental observations.

Stick-slip friction is a well-studied physical phenomenon [11–14], closely related to the fact that static friction coefficients are larger than kinetic friction coefficients. An analogous situation in colloids of charged particles is intermittent shear melting and recrystallization [15–17]. Charge-stabilized suspensions form crystals which can melt by applied shear stress. There is a stick-slip motion associated with transitions between ordered static states and disordered kinetic flow states.

Since it has been established that ER fluids form weak crystals, it is reasonable to expect to see melting transitions and recrystallization in their response to shear. Figure 1 schematically represents the hypothesized behavior. Upon application of the field, an ideal material forms columns of the bct structure as in Fig. 1(a). As shear stress is applied, this structure deforms as in Fig. 1(b). Eventually, the stress is enough to cause a total disruption of the lattice. Earlier experimental work has shown that this occurs in the center of the fluid volume and not at the boundaries. For example, this was verified in the experimental photos of Klingenberg and Zukoski [18]. Once the lattice in the center has been destroyed, the morphology of the ER material is two static solid phases separated by a flowing liquid phase as in Fig. 1(c); the material slips. During the slip some shear stress is released. Eventually, the columnar structure reforms as in Fig. 1(d). A molecular-dynamics-like simulation done

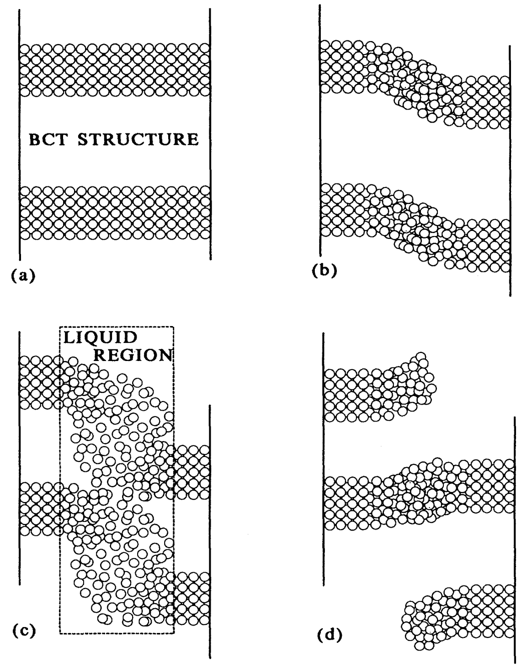

FIG. 1. Schematic diagram of hypothesized shear-induced melting and recrystallizing in an ideal ER fluid.

by Bonnecaze and Brady [6] predicts similar dynamics for ER fluids in an electric field. Their modeled suspension exhibits two distinct motions; "a slow elastic-body-like deformation where electrostatic energy is stored, followed by a rapid microstructural rearrangement where energy is viscously dissipated." Bonnecaze and Brady [19] also determine that the static yield stress is always greater than the dynamic yield stress. If the material exhibits stick-slip response, the relationship of static and dynamic yield stress may be analogous to the relationship between static and kinetic coefficients of friction.

The purpose of this paper is to present experimental evidence of stick-slip response in ER fluids, which is consistent with the above dynamical picture. In Sec. II the experiment is described. In Sec. III the results of this experiment done on a colloid of corn starch suspended in silicon oil are shown. In Sec. IV these results are discussed, in relation to the model of shear-induced phase transitions.

II. EXPERIMENTAL PROCEDURE

In order to carry out our investigation of shearing responses in ER fluids, a small-shear, slow-shear-rate viscometer was built. The apparatus performs three functions: (i) it contains a fixed volume of liquid in a chamber within which the electric field can be varied; (ii) it provides a mechanism to apply a small, known shear to the fluid perpendicular to the direction of the electric field; (iii) it provides a mechanism to measure the strain resulting from the applied stress.

The apparatus [see Fig. 2(a)] consists of a copper cylinder with a leak-proof teflon base. This cylinder is connected to a high-voltage dc power supply. Inside this cylinder is another copper cylinder of smaller radius, which contains a neodymium magnet. The magnet is in the center of the cylinder and its dipole moment is perpendicular to the cylinder's length axis. The base of the inner cylinder (also teflon) has a divot that sits on a pivot point on the base of the outer cylinder. The top of the inner cylinder is copper with a divot in which sits an electrically grounded screw. A small amount of liquid mercury sits in the divot around the grounded screw to insure electrical contact. While containing the ER fluid, this setup allows a voltage to be applied across the two cylinders ($\mathbf{E}=Vd\hat{\mathbf{r}}$). The inner cylinder is free to rotate but is stabilized in the center of the outer cylinder by the

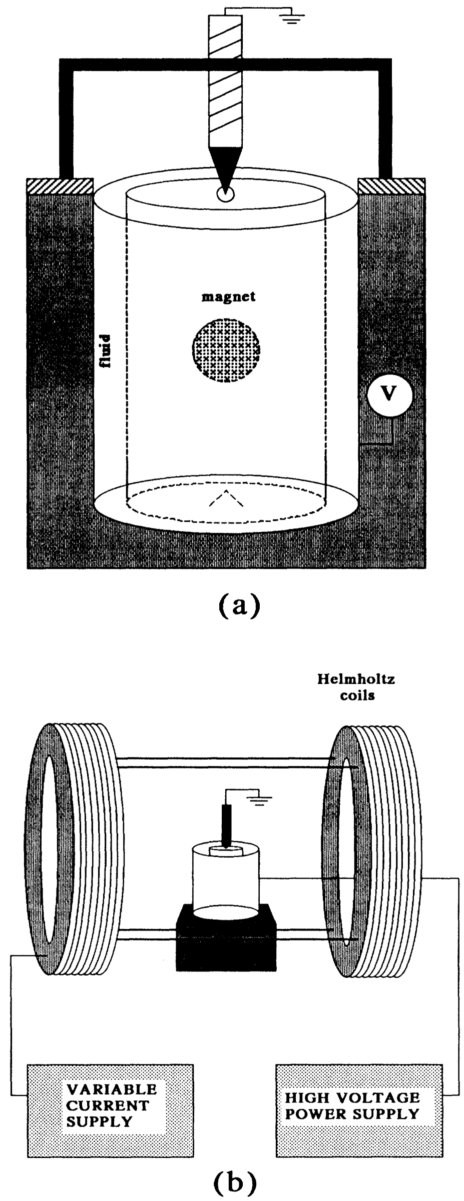

FIG. 2. Sketch of experimental setup: (a) cell used to house the ER fluid, (b) viscometer built for the experiment.

pivot points. This entire set up is placed inside a set of Helmholtz coils [see Fig. 2(b)]. When a current is sent through the coils, a magnetic field is generated. This magnetic field produces a torque on the magnet inside the inner cylinder ($\tau = \boldsymbol{\mu} \times \mathbf{B}$). If a fluid is placed between the cylinders, then a small, known torque can be applied to the fluid while it is in an electric field. A small mirror is attached to the inner cylinder. The deflection of a light beam (measured several m away) provides a measurement of the inner cylinder's rotation angle.

For this experiment, we placed approximately 30 ml of ER fluid between the cylinders. The fluid used was cornstarch (25% by volume) in silicone oil (General Electric SF96-350). The applied voltages shown as examples in this paper include 800 v dc and 400 v dc across a cylinder spacing of 0.4 cm. (The fields were 2.0 and 1.0 kV/cm.)

To collect data about the fluid's mechanical properties at small shear, we performed the following procedure. We turned the electric field on, allowed the fluid to sit undisturbed in the field for at least 10 min—to ensure that the expected microstructure was fully formed—then slowly increased the torque on the inner cylinder. After each small increase in torque, a time interval of approximately 10 min passed and the deflection angle of the inner cylinder was measured. The stress was slowly increased and the strain measured ~ 10 min after each increment until the ER fluid released some of the shear in a quick angular deflection. The process was then continued, increasing the shear stress slowly and measuring the resulting strain.

As the inner cylinder turned to align its internal magnet with the magnetic field, the current required to produce a given torque increased, sometimes exceeding the available supply. At these times, the double cylinder apparatus was rotated with respect to the Helmholtz coils so as to return the magnet to the position perpendicular to the magnetic field ($\tau = \boldsymbol{\mu} \times \mathbf{B}$ is maximum when the angle between $\boldsymbol{\mu}$ and $\mathbf{B}$ is 90°). Of course, this was done slowly and in conjunction with the lowering of the current through the coils so as to maintain the applied torque and not disturb the ER fluid system.

The raw data recorded for a specific applied electric field consisted of each current step through the coils and the corresponding deflection angle of the inner cylinder with respect to its initial position. The linear strain on the material can be approximated by $\sigma = r\theta$. The value of the current through the coils was used to determine the magnetic field and thereby the stress.

III. RESULTS

Figures 3 and 4 show the resulting stress-strain plots. Several facts should be noted. Each data point corresponds to a small increment of increased magnetic field. There is a time interval of ~ 10 min between each. The dotted line shows the applied stress versus the deflection angle during slipping. The slipping occurs with a time scale of ~ 1 sec or less (no data points were actually measured during this time), but an interval of 10 min passed before the next data point was actually measured. Also to be noted is that there is some small, measurable residual torque (τ_0) that is required to move the cylinder even with no field. The source of this torque is the apparatus itself, primarily friction at the pivot points of the inner cylinder. It is a measurable constant of the entire experiment.

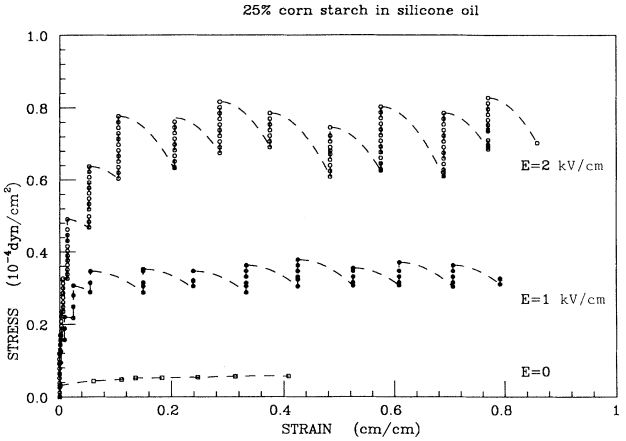

FIG. 3. Plot of applied stress vs resulting strain, showing stick-slip response for a 25%-corn-starch–silicone-oil suspension in electric fields of 0.0, 1.0, and 2.0 kV/cm.

Figure 3 shows the applied torque plotted against the deflection angle of the inner cylinder with the electric field at three different values (2.0, 1.0, and 0.0 kV/cm). For the electric field off, of course the curve is that of a liquid; $\tau = \tau_0$, because liquids support no shear stress. When the electric field is on, the ER fluid resists the shear torque thereby preventing the inner cylinder from turning. Figure 4 shows an expanded view of the initial data of Fig. 3.

For the purpose of discussion, we labeled several features of our data. As mentioned, the torque required to turn the cylinder at zero field was labeled residual torque (τ_0). The phenomenon seen as the stress was slowly increased, sustained by the fluid and then quickly released upon application of only a small increase in stress, we called stick-slip shear response. The average stress at which the fluidlike behavior became dominant

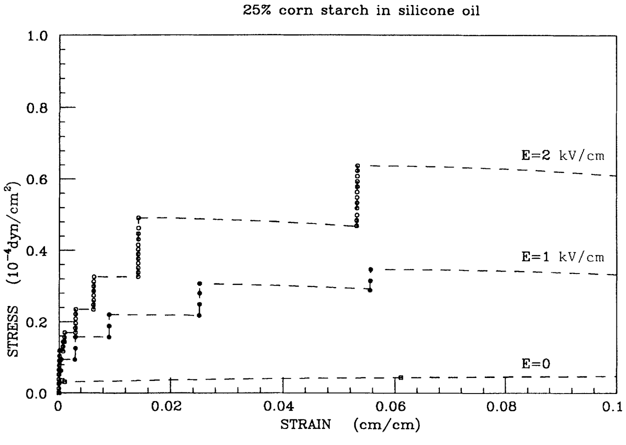

FIG. 4. Expanded view of Fig. 3.

and the material flowed to release the applied stress, we called the static yield stress (τ_{syield}) and claim that it is also the melting stress. While it is true that the material does yield some during the stick-slip shearing, it sustains a significant shear until the yield stress is exceeded. Two other yield stresses are significant. One is the elastic-limit yield stress (τ_{elim}); the stress at which the material suffers a permanent strain that cannot be recovered by removing the stress. From direct observation, the maximum limit for this stress is the stress at which the first slip occurs (see Fig. 4). Also relevant is the dynamic yield stress (τ_{dyield}). While it is not directly tested in this experiment because the applied stress on the material is only increased while the fluid is static, a lower limit on its value can be determined. Obviously, during the intervals of flow, this stress must be exceeded. Equally clear is the idea that when the material stops flowing, its value is no longer exceeded, therefore the average stress at which the material stops flowing is a lower bound on the amount of stress required to keep the fluid flowing.

IV. CONCLUSION

A careful look at the plots reveals several features. As discussed before, it is accepted knowledge that when the electric field is turned on, a microstructure of chained particles is formed. In our experiment, this occurs at zero shear; therefore, the initial structure was formed under static conditions. As we begin to shear this structure (see Fig. 4), we see that it supports a shear, the size of which is dependent on the magnitude of the applied electric field. At some shear, this structure beings to slip, so the inner cylinder makes a small jump. What is interesting is that it stops slipping before much of the applied shear is released. From such observations we may assume that the ER microstructure has not completely broken down, or alternatively, that because the shear is so small, the melted structure can quickly reform. This altered structure, developed under the influence of shear, appears to be stronger in that it can support a further increase in shear. Since this is truly a new structure, it is not surprising that τ_{elim} occurs at this point; release of the shear stress does not return the old structure. Eventually, this structure breaks again and the material flows until it again stops, presumably because another microstructure has formed.

At some applied stress, also dependent on applied electric field, the jumps of the inner cylinder become large (see Fig. 3). During these slips, the electrode-to-electrode columnar structures of the ER microstructure must be disrupted on a larger scale. This is the shear-induced melting discussed earlier. When the inner cylinder stops, it sticks. This must be the result of resolidification. The structures formed after these large slips are similar enough, compared to each other, that it takes approximately the same shear stress to melt them and they result in approximately the same shear strain. A repeatable pattern of stick slip occurs. Since this material is not an ideal ER fluid (it does not have identical spherical particles), this distribution of melting stresses is not surprising because it is practically impossible for the exact same structure to be formed during each resolidification.

From our investigation of the response of this ER fluid to small and slow shearing stresses, we conclude that under these conditions the dynamics of ER fluids demonstrate stick-slip behavior. We argue that shear-induced phase transitions between solid and liquid are the cause of this unique rheology. Further studies probing the experimental time scales of these phase transitions and determining the extent of the melted phase would be useful. Also, experiments with more ideal ER fluids at varying concentrations could be employed to extend this model.

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