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Frequency dependence of electrorheological fluids in an ac electric field

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The frequency dependence of the shear stress of electrorheological (ER) fluid has been studied. We find that the shear stresses of some water-free ER fluids increase monotonically with the frequency and tend to reach saturated values at high frequency. The measurements on $KNbO_3$-silicone-oil ER fluid show that the shear stresses under a 10^3 Hz frequency ac field are several times or even an order of magnitude larger than that under a dc field for the same field strength. The polarization current of the particles in an ac field may strengthen the interaction between them.

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Electrorheological (ER) fluids, which consist of microscopic particles suspended in an insulating liquid, can be transformed into a solidlike state with a dramatic increase in their shear stresses when an electric field is applied. These phenomena have attracted considerable attention [1–6], and many investigations have been carried out on the relationship between the electric field, the structure formation, and the shear stress of ER fluids. However, the effect of the frequency of the applied alternating (ac) electric field on ER fluids has not been sufficiently studied although some inconsistent observations have appeared in the literature [4–10]. In general, they reported a rapid drop or a maximum of the viscosity over a range of ac frequency f from 10 to around 10^3 Hz.

Davis [11–13] analyzed the conductivity effects in ER fluids and concluded that the best performance should be obtained at high frequency for a metal-particle–insulating-oil system, which was demonstrated experimentally by Inoue [14].

In this paper a study of the frequency dependence of the shear stress in an ac electric field is reported. We have found that when an electric field of constant amplitude is applied to some water-free ER fluids the apparent viscosities (or shear stresses) increase monotonically with ac frequency up to 3000 Hz in the range of the instrumental limit. The shear stresses of ER fluids can be several times and even one order of magnitude larger at higher frequency than in a dc field. This may be significant for practical applications of ER fluids. Another subject that arises is what is the physical mechanism of this effect?

According to the conventional expression for the dipole approximation [4,13], for two spherical particles in an ER fluid, the force between the dipoles is

$$f_d \propto p^2 \propto \beta^2 , \tag{1}$$

where p is the dipole moment and β is a factor given by

$$\beta=\frac{\tilde{\epsilon}_p(\omega)-\tilde{\epsilon}_f(\omega)}{\tilde{\epsilon}_p(\omega)+2\tilde{\epsilon}_f(\omega)} . \tag{2}$$

The complex dielectric constant $\tilde{\epsilon}=\epsilon+\sigma/i\epsilon_0\omega$, σ is the conductivity, and the angular frequency $\omega=2\pi f$. The subscripts p and f indicate the particle and the fluid, respectively. We then have

$$\beta^2=\frac{(\sigma_p-\sigma_f)^2+\omega^2\epsilon_0^2(\epsilon_p-\epsilon_f)^2}{(\sigma_p+2\sigma_f)^2+\omega^2\epsilon_0^2(\epsilon_p+2\epsilon_f)^2} . \tag{3}$$

It is obvious that, when the condition $|\epsilon_p-\epsilon_f|/(\epsilon_p+2\epsilon_f)>|\sigma_p-\sigma_f|/(\sigma_p+2\sigma_f)$ is satisfied, β^2 should monotonically increase with increasing frequency and reach $[(\epsilon_p-\epsilon_f)/(\epsilon_p+2\epsilon_f)]^2$ at high frequency, i.e., the ER fluid shear stress, which is proportional to the force between the dipoles, should be enhanced by increasing the frequency of the ac field and should tend to a saturated value if the frequency is high enough.

In experiments, for measuring ER fluids, the electrodes with ER fluid in between constitute a capacitor in the ac electric circuit. The voltage between the electrodes and the current passing through the capacitor are $U=U_0\cos\omega t$ and $I=I_0\cos(\omega t+\varphi)$, respectively. For an ideal capacitor, $\varphi=\pi/2$ and $I_0=U_0\omega C$, where C is the capacity. In the ac high voltage measurement usually a transformer is utilized which behaves as an equivalent inductance. Therefore the measurement can be simulated by a complex circuit with capacitance, inductance, and resistance. As the input voltage of the power supply is fixed the voltage between the ER fluid electrodes must vary with the frequency. A maximum voltage will appear on the electrodes at the resonance frequency, above which the voltage will decrease and become very low at higher frequencies. The voltmeter used in the measurement should have high enough input impedance and be frequency independent in the measured range. In order to keep a constant magnitude of the electric field between the electrodes it is necessary to adjust the voltage of the power supply carefully when changing the frequency.

A high voltage ac power supply was designed and set up for the experiment. The sinusoidal voltage output and its frequency are continuously adjustable, their effective ranges being 0–5 kV and 10 Hz–40 kHz, respectively. However, in most of the measurements on ER fluids reports herein the current output limited the frequency to be applied to about 3 kHz with high voltage output. Beyond that frequency the power supply is unable to withstand the large current caused by the capacitance effect. The bob and cup of the rheometer filled with ER

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fluid constitute a capacitor of capacity C. When only pure silicone oil is used, then $C \simeq 180$ pF, and the capacitor is almost ideal, with a phase shift of $\varphi = \pi/2$ which changes very little in the ranges of the frequency and voltage measured. By measuring the voltage across a resistance $R = 1$ kΩ in series with the capacitor, which is about three orders of magnitude smaller than the impedance of the ER fluid in the measured frequency range, we calculated the current passing through the ER fluid using a Keithley 191 digital multimeter with 10 MΩ input impedance. The voltage across the ER fluid was measured with a digital multimeter with 1000 MΩ input impedance.

The frequency dependences of the shear stresses in several kinds of ER fluids have been studied. The ER fluids consist of silicone oil with suspended $KNbO_3$, $BaTiO_3$, $SrTiO_3$, $LiNbO_3$, or Si particles. All particles were prepared by grinding the single crystals and then drying in an oven above 200 °C to evaporate any water. As an example, we present the results on the $RNbO_3$–silicone-oil ER fluid.

Particles of a ferroelectric $KNbO_3$ crystal (average size 35 μm), the permittivity of which was measured at room temperature to be $\epsilon_p \simeq 100$, were suspended in silicone oil. The volume fractions were 0.025, 0.05, 0.075, and 0.1, respectively. The rheological measurements were performed on a rotating cylinder rheometer.

If the voltage of the power supply is fixed, the mean effective voltage across the ER fluid will vary with the frequency and will reach a maximum at a resonance frequency $f_r = [LC]^{-1/2}$, where L is the equivalent inductance and C the capacity in the circuit. Correspondingly, a maximum of the shear stress will be observed at that resonance frequency. Generally, f_r is in the range of a few Hz to a few kHz. It is incorrect to measure the output voltage of the power supply instead of the voltage across the ER fluid; this may be the reason why some previous authors observed a maximum or rapid decrease of the shear stress at higher frequency when the ac electric fields were applied. Even a negative peak on the shear stress curve as reported earlier [7] can be observed in some cases with an unsuitable circuit. Therefore care must be taken in experiments, with ac field, especially for the voltage measurement.

When we keep a fixed mean effective voltage U_c across an ER fluid at different frequencies by adjusting the output of the power supply, the mean effective current I passing through the ER fluid follows a linear rule with respect to the frequency, i.e., $I = U_c \omega C$. All the following results were measured with the actual electric field across the ER fluid instead of the output of the power supply. Figures 1(a) and 1(b) show the frequency dependences of the currents and the shear stresses, respectively, for different electric fields. Figures 2(a) and 2(b) show how the currents and the shear stresses change with the field strength at different frequencies.

The currents vary linearly with the frequency at a fixed field but not linearly with the electric field at a fixed frequency. These facts indicate that the equivalent permittivity of the ER fluid does not change with the frequency in the measuring range but does change with the electric field, i.e., the relation between the current and electric field no longer follows the Ohmic rule as normally observed in a dc electric field. One reason is that rearrangement in the suspensions, such as formation of chains and columns due to the higher field, causes the permittivity of the ER fluid to increase. A detailed explanation of this effect will be reported elsewhere.

Although the tendency of the shear stress increasing monotonically with the frequency at a fixed field can be predicted with the analysis described in Eqs. (1)–(3), a quantitative calculation consistent with experiment still cannot be given. If we calculate β^2 by using Eq. (3) with parameters $\sigma_p = 10^{-8}$ S/m, $\sigma_f = 10^{-7}$ S/m, $\epsilon_p = 100$, and $\epsilon_f = 2.5$, the dashed line plotted in Fig. 1(b) fits the measured curve well. However, the measured conductivity of the silicone oil, σ_f, is 3.45×10^{-13} S/m; with this value the calculation, which gives a very slow increase with the

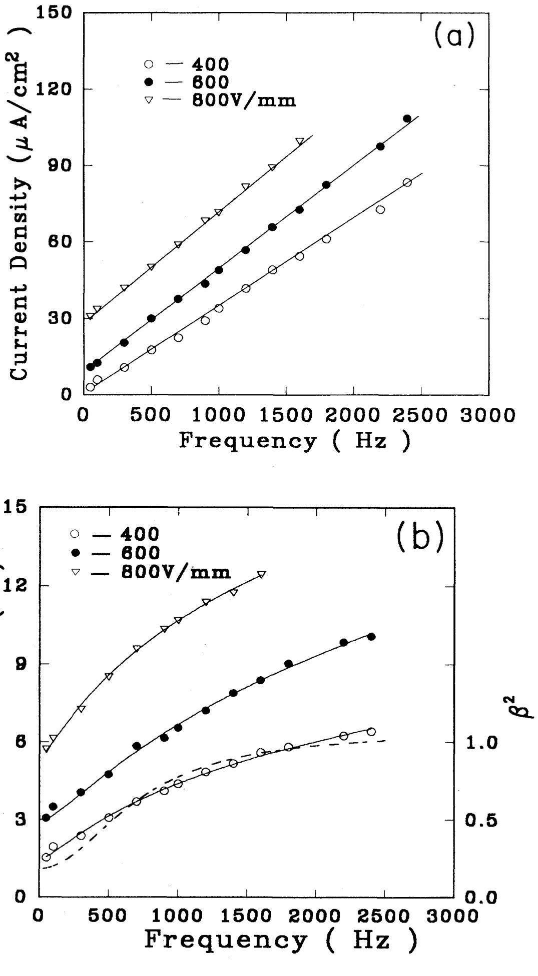

FIG. 1. Variation of the mean effective current (a) and shear stress (b) of $KNbO_3$–silicone-oil ER fluid ($\phi = 0.1$) with frequency for $E_c = 400$, 600, and 800 V/mm, respectively (shear rate 15.50 s^{-1}); dashed line is β^2. The curve for β^2 is obtained from Eq. (3) with $\epsilon_p = 100$, $\epsilon_p = 2.5$, $\sigma_p = 10^{-8}$ S/m, and $\sigma_f = 10^{-7}$ S/m.

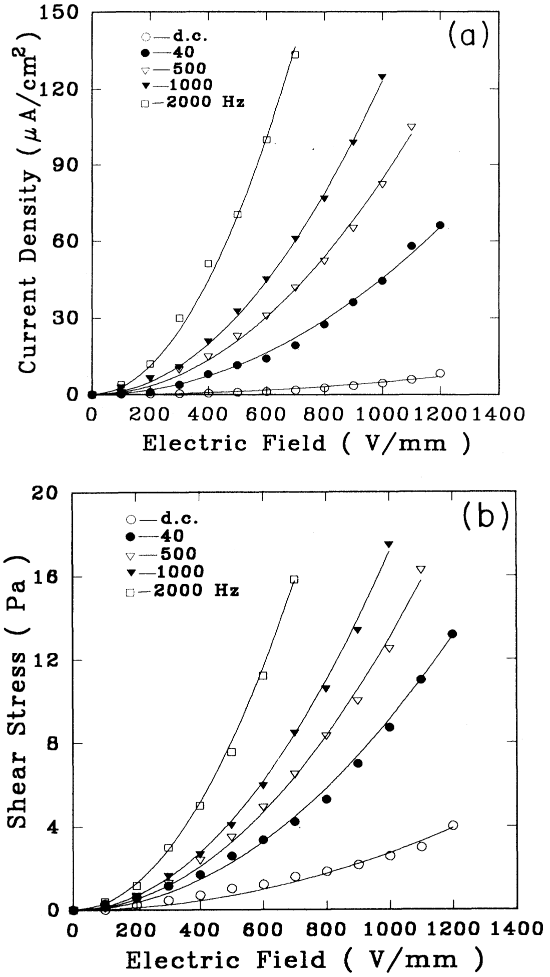

FIG. 2. Variation of the mean effective current (a) and shear stress (b) of $KNbO_3$-silicone-oil ER fluid ($\phi=0.1$) with the mean effective electric field for $f=0$, 40, 500, 1000, and 2000 Hz, respectively (shear rate 15.50 s^{-1}).

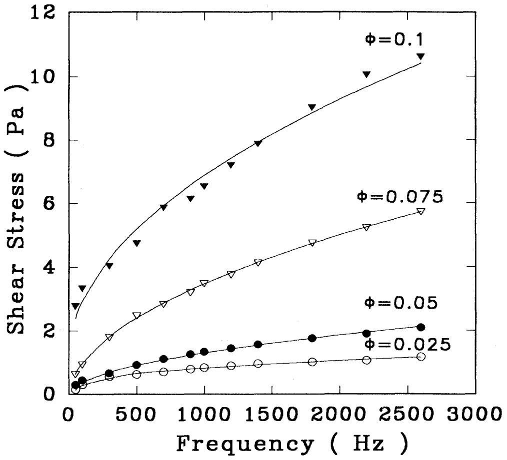

FIG. 3. Variation of shear stress of $KNbO_3$–silicone-oil ER fluid with frequency for different volume fractions $\phi=0.025$, 0.05, 0.075, and 0.1, respectively ($E_c=600$ V/mm; shear rate 15.50 s^{-1}).

frequency, cannot fit the measured result well. The reason for the disagreement is not understood.

On the other hand, the electric field dependence of the shear stress at a certain frequency is the same as that in the case of a dc field. However, it can be seen that in Fig. 2(b) the shear stresses at a higher frequency are several times or even an order of magnitude larger than that under a dc field for the same field strength. For instance, at 700 V/mm, the shear stress is 16.6 Pa at $f=2000$ Hz but 1.6 Pa in a dc field.

The frequency dependences of the shear stresses for different volume fractions $\phi=0.025$, 0.05, 0.075, and 0.1 have also been measured and are shown in Fig. 3. The behavior of the shear stress increasing with ϕ at a fixed frequency is about the same as that in a dc field.

In addition to the $KNbO_3$–silicone-oil ER fluid, $BaTiO_3$, $SrTiO_3$, $LiNbO_3$, or Si particles suspended in silicone oil exhibited the same phenomena, that is, the shear stresses increased monotonically with the frequency up to 3 kHz. We also measured systems of Al_2O_3 and SiO_2 particles suspended in silicone oil. It is well known that the ER response of dry Al_2O_3 and SiO_2 particles is rather poor, so the frequency dependences were hardly detectable.

All of our results were obtained with anhydrous ER fluids. In the case of water-activated ER fluids the frequency responses greatly depend on the water content and usually a maximum of the shear stress appears in the frequency range below 10^3 Hz [15]. This was also observed in our experiments on some ER fluids with water-mixed suspensions, and must be due to the dielectric and surface-active properties of the water on the particles, as discussed by many authors. As for the Si–silicone-oil fluid, the behavior may be like that of the metal-particle–oil system discussed by Davis [13].

In conclusion, we have found that the shear stresses of some water-free ER fluids increase monotonically with the frequency and tend to reach saturated values at high frequency under a fixed ac field strength. This phenomenon depends on the mismatch of the conductivity and the permittivity of the particle and the fluid. As an example, detailed results of the $KNbO_3$-silicone fluid are reported in which remarkable enhancement of the shear stresses has been observed in the frequency range 10 Hz–3 kHz compared with that in a dc field. The frequency response of ER fluids indicates that under an ac electric field the polarization current of ER fluids which increases with frequency must strengthen the interaction between the particles. A further study on the mechanism of the frequency effect in ER fluids will be made.

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