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Electrorheological resonance observed in a colloidal suspension

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Measurements of rheological properties of a suspension containing rutile (TiO_2) particles are made under applications of ac electric fields. When the shear stress is measured as a function of frequency of the ac field, a peak in the shear stress is observed with the peak frequency shifting linearly with the shear rate. The effects of the shear rate and the amplitude of the field on the peak indicate that the peak is due to a resonance that occurs when the angular velocity of rotating particles matches the angular frequency of the ac field. The resonance makes this suspension behave as a non-Newtonian fluid.

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I. INTRODUCTION

When an electric field of a few kV mm^{-1} is applied to a suspension consisting of particles of a few micrometers and electrically nonconducting liquid, the apparent viscosity of the suspension increases. This effect is called an electrorheological (ER) effect and was first found by Winslow [1] in particulate suspensions containing water adsorbed particles. Owing to a dielectric mismatch between the particle and the liquid, an electric field induces a polarization at the interface between the particle and the liquid. Interactions between these polarizations cause chain and/or column structures in the suspension and give rise to a large increase in the apparent viscosity [2–4].

In the absence of the electric field there occurs a rotation of the particles under a steady shear flow, especially when the suspension is dilute. This behavior is due to a viscous torque exerted on the particle and theoretically investigated by Jeffrey [5]; the rotational motion is characterized by an angular velocity of (shear rate)/2 if the particles are spherical. This is confirmed by Block *et al.* [6] from the measurements of the dielectric constant under a shear flow, i.e., flow modified permittivities. When an electric field is applied to the suspension, the rotational motion of the particles is modified. Some theoretical works [7–10] have shown that the induced polarization and the electric field exert an electric torque on the particles to suppress the rotational motion of the particles, and this effect also contributes to an increase in the viscosity. The fundamental studies of the ER effect have just begun and there are many factors to be clarified. The rotational motion of the particles is an important factor to be investigated, since it is strongly associated with the ER properties [11,12]. However, its behavior under the electric field and its effect on the ER effect are not experimentally clarified.

In the present study electrorheological properties of an anhydrous suspension containing rutile (TiO_2) particles are investigated. At room temperature, rutile has a tetragonal crystal structure and large static dielectric constants of 170 along the c axis and 86 along the a axis [13]. In a suspension consisting of rutile particles and nonconducting oil, e.g., silicone oil ($\varepsilon_l \sim 2$), we can expect an appearance of the induced polarization responsible for the ER effect, since an electric field induces a polarization along the field with its amplitude given by $4\pi\varepsilon_l a^3[(\varepsilon_p-\varepsilon_l)/(\varepsilon_p+2\varepsilon_l)]E$, where ε_p is a dielectric constant of the particle, ε_l a dielectric constant of the liquid, a a radius of the particle, and E an electric field. However, the ER effect of this suspension is small and cannot be understood from the conventional theory of the interfacial polarization [14], which motivates us to examine the ER effect of this suspension. In the course of our studies on the relationship between the polarization and the ER effect of this suspension, we observed a peak in shear stress when the shear stress was measured as a function of the frequency of the ac electric field. The effects of the shear rate and the amplitude of the field on the peak indicate that the observed peak is due to a resonance that occurs when the angular velocity of the rotating particles in the steady flow matches an angular frequency of the electric field, and we call this phenomenon ER resonance. It is also found that the resonance causes the rheological property of the suspension to behave as a non-Newtonian fluid.

II. EXPERIMENT

The present experiments employed a 5.5 vol % suspension prepared by mixing rutile (TiO_2) particles of average diameter 0.4 μm (TM-1, Fuji Titanium Co., Ltd., Japan) with a silicone oil of viscosity 50 cP (TSF451-50, Toshiba Silicone Co., Ltd., Japan). The shape of the particles was not spherical but irregular, with some facets. To obtain an anhydrous suspension, the particles were dried by keeping the materials for six hours under a condition of 10^{-6} torr and 160 °C, and silicone oil was dehydrated using molecular sieves 4 A.

Rheological properties were measured at 300 K using a homemade concentric cylindrical viscometer, which

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operates in the shear rate region of 6.59 to 2000 s^{-1}. To apply an electric field perpendicular to the flow, the inner and outer cylinders were electrically insulated to enable the application of the high voltage across the 1 mm gap between these cylinders. For generating ac high voltages, a variable frequency ac power source (CVFT1-50HVP2, Tokyo Seiden Co., Ltd., Japan) was used. The frequency dependence of the ER effect under applications of the ac field was measured except for the region centered around 50 Hz, since at this region a forced vibration occurred in our viscometer. Throughout this article the amplitude of the ac field is expressed in rms.

III. RESULTS AND DISCUSSION

Figure 1 shows the results of the shear stress vs the frequency of the ac field when the shear rate is varied. The amplitude of the field is 2 kV mm^{-1}. As is evident from the figure, a peak is observed in the shear stress whose position and shape are dependent on the shear rate. With an increase in shear rate, the peak frequency shifts to higher values and the shape of the peak becomes broader, indicating that the origin of the peak is associated with the shear rate. In Fig. 2 the peak frequency ν_{peak} is plotted against the shear rate $\dot{\gamma}$, showing that the peak frequency increases linearly with the shear rate with a relationship of $\nu_{\text{peak}}=0.066\times\dot{\gamma}$.

For clarification of the origin of the peak, the experimental study of the flow modified permittivity performed by Block *et al.* [6] is suggestive. They measured the frequency dependence of dielectric constants of a suspension consisting of latex particles and heptane under a steady shear flow, finding a peak in the dielectric permittivity. When the shear rate is increased, the position of the peak shifts to higher frequencies and its shape becomes broader. The peak frequency f_{peak} is shown to be proportional to the shear rate $\dot{\gamma}$ with a relationship of $f_{\text{peak}}\approx\dot{\gamma}/4\pi$. Concerning the motion of the spherical particles under a steady shear flow, it is theoretically shown that the angular velocity of the rotating particles is given by $\dot{\gamma}/2$ [5,7,10]. On the basis of these results, the origin of the peak is attributed to a resonance caused by a coupling between electric field induced charge displacement and forced rotation of the particles under the steady shear flow.

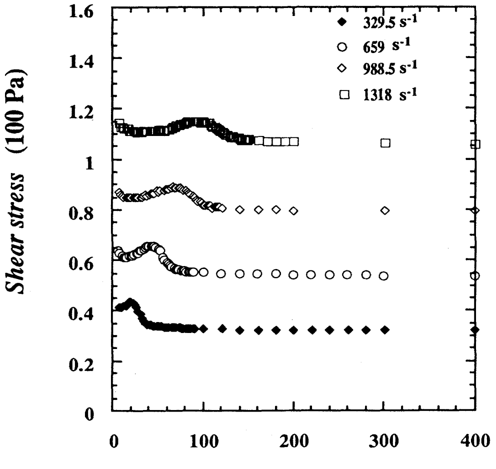

FIG. 1. Shear stress vs frequency of the ac field (2 kV mm^{-1}) in a suspension consisting of 5 vol % rutile particles and silicone oil. A peak in the shear stress is observed, with the position and the shape depending on the shear rate.

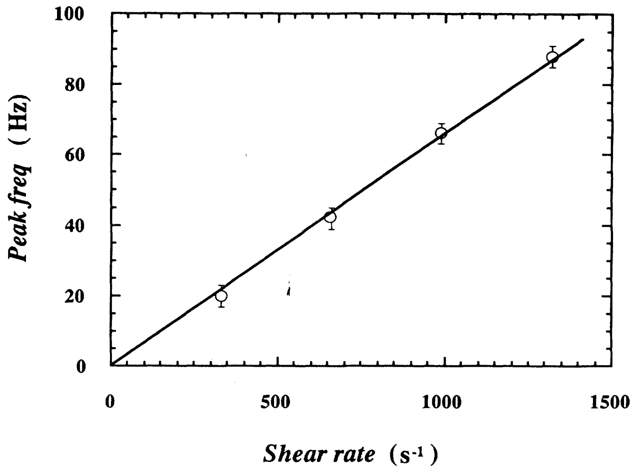

FIG. 2. Peak frequency ν_{peak} vs shear rate $\dot{\gamma}$ obtained from Fig. 1. ν_{peak} increases linearly with $\dot{\gamma}$ with a relationship of $\nu_{\text{peak}}=0.066\dot{\gamma}$.

These results suggest that the peak observed in the shear stress (Fig. 1) is also due to a resonance associated with the rotational motion of the particles. The obtained proportional constant 0.066 is smaller than the value $1/4\pi(\approx 0.080)$ expected from the rotational motion of the particles in no electric field. This difference, however, can be understood since the smaller values of the proportional constant can be expected if we consider a suppressed rotational motion of the particle under the high electric field of 2 kV mm^{-1}. Some theoretical works [7–10] have suggested that when a large amplitude of the electric field is applied, the rotational motion of the particles is modified, because of a torque caused by the induced polarization and the electric field and/or the dipolar interactions between the particles. To make clear whether the smaller proportional constant is due to the suppressed rotational motion or not, the frequency dependence of the shear stress is observed as a function of the amplitude of the ac field (Fig. 3). At low electric fields less than c.a. 1 kV mm^{-1}, a distinct peak is not observed because of the lower resolution of our instrument in a small shear stress region. With an increase in the amplitude of the electric field, the peak becomes clearly observable, with its peak position shifting to lower frequencies. This can be understood as a result of the suppression of the rotational motion at higher electric fields. The solid line in this figure shows a shift of the peak position when the amplitude of the ac field is varied, which shows the noticeable result that the peak frequency at the limit of zero electric field converges to $659/4\pi=52.4$ Hz—which is just the value theoretically expected for the rotational frequency of the particles in

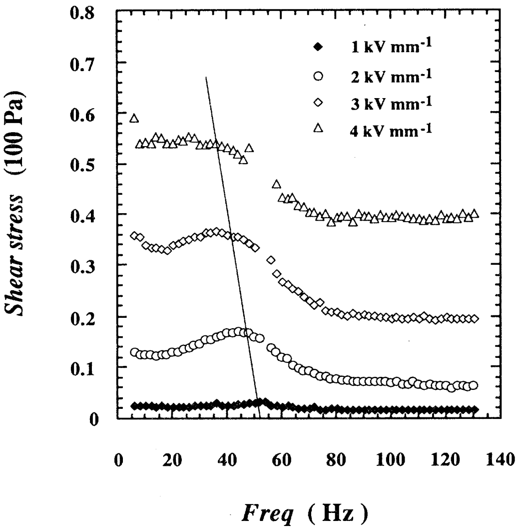

FIG. 3. Effect of the ac field amplitude on the peak at a shear rate of 659 s^{-1}. The solid line indicates a change of the peak frequency when the amplitude of the field is varied. At the limit of zero field, the peak frequency converges to $659/4\pi = 52.4$ Hz, which is the value theoretically expected for the rotational motion under no electric field.

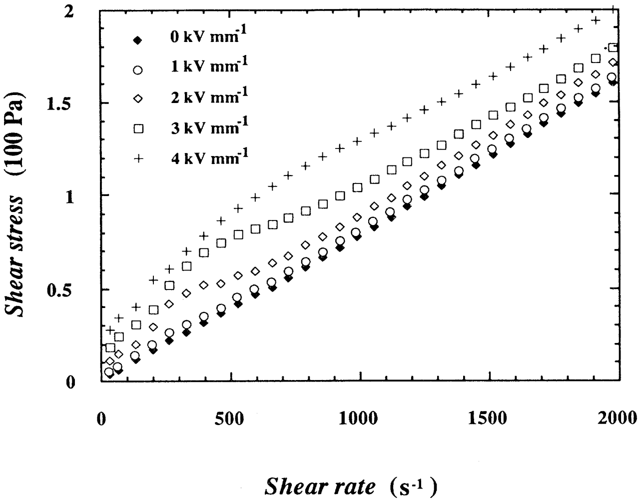

FIG. 4. Shear stress vs shear rate when the amplitude of the ac field (20 Hz) is varied. With an increase in the amplitude, the suspension behaves as a non-Newtonian fluid with a hump in the shear stress, which can be interpreted from the ER resonance.

the case of no electric field. These results furthermore indicate that the peak is due to a resonance and suggest following particle motion in the suspension. If the electric field is not applied, the particles under a steady shear flow are rotating with an angular velocity of $\dot{\gamma}/2$. The application of the electric field changes the particle motion; the angular velocity of the rotation decreases because of the suppression of the rotational motion caused by the electric torque.

The effect of the resonance is reflected in the rheological property of the suspension. Figure 4 shows a result of the shear rate dependence of the shear stress when the amplitude of the electric field is varied. In the case of no electric field, the suspension behaves as a Newtonian fluid, as shown by the fact that the shear stress increases proportionally to the shear rate. When an ac electric field of 20 Hz is applied, a hump appears in the shear stress to make the suspension a non-Newtonian fluid. With an increase in the amplitude of the electric field, the hump appears at a higher shear rate, which can be understood from the relationship of $\dot{\gamma} = \nu_{\text{peak}}/C$, given in Fig. 2, since the increase in the amplitude of the electric field makes the proportional constant C smaller and hence makes the resonance occur at higher shear rate.

Although a thorough understanding of the above mentioned ER resonance will occur in the future, some theoretical works [10,15] of the induced polarization on a rotating particle may give a qualitative insight into this effect. Provided that a spherical particle is rotating about the x axis with an angular velocity Ω under the application of an electric field $E_0 \cos\omega t$ along the z axis, polarizations are induced along the y axis as well as along the z axis [Eqs. (1) and (2)] [15]:

$$m_y(t) = E_0(\alpha_0 - \alpha_\infty)\Omega\tau \,\text{Re}\{(e^{i\omega t}/[1-(\omega^2-\Omega^2)\tau^2 + 2i\omega\tau]\} \ , \tag{1}$$

$$m_z(t) = E_0(\alpha_\infty \cos\omega t + (\alpha_0 - \alpha_\infty)\text{Re}\{e^{i\omega t}(1+i\omega\tau)/[1-(\omega^2-\Omega^2)\tau^2 + 2i\omega\tau]\}) \ . \tag{2}$$

Here τ is a correlation time of the polarization and α_0 and α_∞ are polarizabilities at $\omega = 0$ and $\omega \gg \tau^{-1}$, respectively. If $\omega\tau > 1$, singularities appear in these polarizations when ω approaches Ω; m_y has a maximum and a minimum below and above Ω, respectively, and m_z has a maximum at Ω. In the present case the particle (rutile) has a conductivity of less than 10^{-12} S cm^{-1} and a dielectric permittivity of 170 [13,16], leading to the correlation time τ longer than 1 s if we consider that the polarization is of interfacial type with its correlation time given by ε/σ. This magnitude of τ fulfills the condition of $\omega\tau > 1$, and the singularities are expected in the polarizations. Of these polarizations the y component exerts a torque on the particle in the electric field along the z axis, and the z component generates dipolar interactions between the particles, making the angular velocity Ω lower than $\Omega_0 = \dot{\gamma}/2$, especially when ω approaches Ω. Considering the theoretical result that the contribution of the suppressed rotational motion to the viscosity is proportional to $(\Omega_0 - \Omega)$ [7–10], we can expect a peak in the shear stress when $\omega \approx \Omega$. It should be mentioned that such an ER resonance would not appear in all the ER suspensions; in the system where the dipolar interaction between the particles is strong, the resonance will not be expected since the particle rotation is prevented as a result of the formation of a chainlike cluster. In the present experiment we employ a suspension containing nonspherical particles having a size distribution, which may result in a broadening of the resonance peak. Measurements on a suspension of monodisperse spherical particles would more clearly shed light on this ER resonance.

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[1] W. M. Winslow, J. Appl. Phys. **20**, 1137 (1949).

[2] A. P. Gast and C. F. Zukoski, Adv. Colloid Interface Sci. **30**, 153 (1989).

[3] D. J. Klingenberg, F. van Swol, and C. F. Zukoski, J. Chem. Phys. **91**, 7888 (1989).

[4] H. Block and J. P. Kelly, J. Phys. D **21**, 1661 (1988).

[5] G. B. Jeffrey, Proc. R. Soc. London, Ser. A **102**, 161 (1922).

[6] H. Block, K. M. W. Goodwin, E. M. Gregson, and S. M. Walker, Nature (London) **275**, 633 (1978).

[7] A. Okagawa, R. G. Cox, and S. G. Mason, J. Colloid Interface Sci. **47**, 536 (1974).

[8] A. Okagawa and S. G. Mason, J. Colloid Interface Sci. **47**, 568 (1974).

[9] H. Brenner, J. Colloid Interface Sci. **32**, 141 (1970).

[10] J. Hemp, Proc. R. Soc. London, Ser. A **434**, 297 (1991).

[11] H. Block and J. P. Kelly, J. Phys. D **21**, 1661 (1988).

[12] K. Negita and Y. Ohsawa, J. Phys. (France) II (to be published).

[13] R. A. Parker, Phys. Rev. **124**, 1719 (1961).

[14] F. Filisko, in *Electrorheological Fluids: Mechanics, Properties, Structure, Technology and Applications,* edited by R. Tao (World Scientific, Singapore, 1992), p. 116.

[15] H. Block, E. Kluk, J. McConnell, and B. K. Scaife, J. Colloid Interface Sci. **101**, 320 (1984).

[16] D. C. Cronemeyer, Phys. Rev. **87**, 876 (1952); L. E. Hollander, Jr., and P. L. Castro, J. Appl. Phys. **33**, 3421 (1962).