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Experimental investigation of the frequency dependence of the electrorheological effect

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The frequency dependence of the electrorheological response was studied experimentally in a suspension of barium titanate spherical particles suspending in silicone oil. In the system, only one factor, namely the frequency of the applied electric field, affects the electrorheological effect. The experimental data reflect the frequency effect more reliably and more accurately. Under the sinusoidal electric fields, the shear stress increases sharply with frequency below 500 Hz and reaches a saturated value beyond 500 Hz. The phenomena can be explained well with the permittivity mismatch theory. More experiments indicate that the electrorheological effect should be the sum of the mismatch polarization and the interfacial polarization.

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I. INTRODUCTION

Small solid particles suspended in an insulating liquid always arrange themselves into chains, and subsequently into columns under an external electric field. At the same time, the rheological properties, such as viscosity and shear stress of the two-phase suspension, change greatly and the suspension shows a solidlike behavior at high electric fields [1]. These phenomena have attracted considerable attention due to the potential applications in the industry. The rheological behavior is generally attributed to the formation of particle chains or particle columns formed in the insulating liquid, which are induced from the interface polarization (double-layer model [1,2]) or from the volume polarization (permittivity mismatch model [3,4]) of the particles under electric fields.

The double-layer model [1,2] supposes there is a double electric layer around the solid particles which are suspended in an insulating liquid. Under electric fields, the double layer is polarized, deformed, and elongated, and the polarization dominates the electrorheological (ER) effect. The model is very successful in surfactant-activated ER systems, especially in water-activated systems. However, after the appearance of water-free or so-called surfactant-free ER fluids, the model is replaced by a permittivity mismatch model which supposes that the volume polarization, and not the interface polarization, dominates the ER effect.

According to the mismatch model [3,4], the shear stress is proportional to the square of the dipole moment $\vec{P}$ of the particles. The induced dipole moment $\vec{P}$ of a uniform spherical particle with a complex dielectric permittivity $\tilde{\epsilon}_p(\omega) = \epsilon_p(\omega) + \sigma_p / i\epsilon_0\omega$ [$\epsilon_p(\omega)$ and σ_p are the relative dielectric constant and electric conductivity of particles], which is suspended in a liquid with a complex permittivity $\tilde{\epsilon}_f(\omega) = \epsilon_f(\omega) + \sigma_f / i\epsilon_0\omega$, can be expressed by

$$\vec{P}(\omega) = 4\pi\epsilon_0 r^3 \mathrm{Re}[\tilde{\epsilon}_p(\omega)]\beta(\omega)\vec{E}_{\mathrm{local}}(\omega), \tag{1}$$

where r is the radius of the particles, $\vec{E}_{\mathrm{local}}(\omega)$ is the local electric field, and $\beta(\omega)$ is the mismatch factor, which is given by [5]

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

and

$$|\beta(\omega)|^2 = \frac{(\sigma_p - \sigma_f)^2 + \omega^2\varepsilon_0^2[\varepsilon_P(\omega) - \varepsilon_f(\omega)]^2}{(\sigma_p + 2\sigma_f)^2 + \omega^2\varepsilon_0^2[\varepsilon_p(\omega) + 2\varepsilon_f(\omega)]^2}. \tag{3}$$

It is concluded that the shear stress of ER fluids is dependent on the permittivity of the particles, the permittivity of the liquid, the amplitude of the external electric field, and the particle size, as well as on the angular frequency $\omega = 2\pi f$ of the applied electric fields.

Many experiments have been carried out to study the rheological dependence on the permittivity [2,6,7], on the amplitude of the electric fields [1], and on the particle size [8]. Agreements have been obtained between experiments and theory. However, the frequency dependence of ER properties is not very clear because of complex experimental conditions. The observed phenomena are contradictory. For example, in some water-free ER systems, the shear stress increases monotonically with frequency [5,9,10], while in most of the surfactant-activated systems, especially water-activated systems, the shear stress decreases with frequency [11]. Another complex relationship has also been reported in the literature [12]. Generally, the dissimilarity is attributed to the surfactants on the particles. However, even in the surfactant-free systems, different ER fluids show different frequency behaviors. In the glass particle/vacuum pump oil water-free systems, the shear stress decreases with frequency [13]. In the zeolite/silicone oil systems, the stress is nearly a constant with increasing frequency [14]. In $KNbO_3$/silicone oil ER fluids, the shear stress increases with frequency [5]. Up to now, no systematic explanation has been given on the frequency dependence of the ER effect. Therefore, the frequency dependence of the ER effect has not been sufficiently understood.

The particle materials used in ER fluids are insulating ceramic powders, semiconductors, metal or polymers, or even core-shell structured particulates [1,15]. The permittivity of the materials is usually complex, depending on the frequency of the external alternating electric field [1], as is the liquid permittivity. When we vary the frequency of the external electric fields to measure the frequency dependence of the ER effect in experiments, the frequency not only influences the ER effect directly, but it also changes the particle permittivity, which also controls the ER effects. From Eq. (3) and the experimental studies [6,7,16,17], it is very obvious that the particle permittivity has great influences on the rheological characteristics of ER fluids. Therefore, the frequency dependence of the particle permittivity makes the experimental investigations on the frequency dependence of the ER effect very difficult. In the reported literature, the measured frequency dependence of the ER effect usually contained both the frequency-induced permittivity contribution and the direct frequency contribution to the ER effect. From experiments, we cannot distinguish which parameter (permittivity or frequency) causes the decrease or increase of the shear stress and how the shear stress increases or decreases with frequency. This should be one of the important reasons why the contradictory results have been observed on the frequency dependence. Therefore, in order to understand the unequivocal relationship between frequency and the ER effect, an ideal electrorheological system, in which particle permittivity is frequency-independent, should be employed.

In this paper, we choose barium titanate/silicone oil systems as ER fluids to carry out an experimental study of the frequency dependence of the shear stress, in which both barium titanate $(BaTiO_3)$ permittivity and silicone oil permittivity are independent of frequency. In order to avoid the effect of the interfacial polarization, the system is surfactant-free. Under our experimental conditions, only one factor, namely the frequency of the applied alternating electric (ac) field, varies while all the other parameters, such as the permittivity and the electric fields, etc., are constant. The experimental data should reflect the more reliable and more accurate relationship between the frequency of electric fields and the ER effect.

II. EXPERIMENT

ER fluids consisted of spherical $BaTiO_3$ single-crystal particles suspended in silicone oil. $BaTiO_3$ single crystals were ground to particle powder, sieved, and then thermally sprayed. During the thermal spray, the outlayer of the irregular particles was semimelted under high temperature and the particles became spherelike because of surface tension. The obtained spheres were single crystals [18]. After being thermally sprayed, the particles were washed with de-ion water and sieved again. The average diameter of the particles was 35 μm. After being dried at 110 °C in vacuum to evaporate any water, the particles were mixed with silicone oil. The silicone oil was boiled in vacuum for 30 min before being mixed with the particles in order to minimize the absorbed water.

The dielectric measurements were carried out on a HP4192A impedance analyzer. $BaTiO_3$ single-crystal powders were compacted into a disk. Metal Au was evaporated on opposite faces as electrodes. After measuring their capacitance and loss angle ($\tan \theta$), the dielectric constant and ac conductivity were calculated. When measuring the dielectric constant and electric conductivity of the silicone oil, the silicone oil was filled into a capacitor consisting of two parallel electrodes. The details were described in Ref. [19]. Because of the limit of the impedance analyzer, the dielectric measurements were carried out only at low voltage (~ 1 V).

The rheological measurements were performed on a modified NXS-11 model rotating cylinder rheometer (Chengdu, China). ER fluids were poured into the gap between the bob and the cup of the rheometer. The whole sample system was immersed in an oil bath where the temperature was controlled at room temperature. The details were described in Ref. [19].

In experiments to measure the shear stress of ER fluids under electric fields, a self-designed high-voltage ac power supply was used. The sinusoidal voltage output $U = U_0 \sin(\omega t)$ and its frequency $\omega = 2\pi f$ were continuously adjustable. It provided a 5–2000 Hz sinusoidal wave across ER samples, of peak amplitude U_0 ranging from 0 to 5000 V. The dc biasing voltage was zero. When the frequency was changed, the voltage amplitude U_0 applied on ER fluids usually varied [5] because of the resonance between the power supply and the capacitor consisting of the bob and the cup of the rheometer. Simultaneously, we adjusted the power supply to hold the voltage amplitude on ER fluids. At high frequency and high voltage, the sinusoidal waveform was always distorted into a trapezoid waveform. In order to apply a sinusoidal wave on ER fluids, an ac/dc oscilloscope (Sanke HZ4318, China) was used to monitor the waveform of the ac electric field, and to measure the electric field amplitude simultaneously. The dc electric field (frequency is zero) was provided by a dc power supply (Beijing, China).

III. RESULTS AND DISCUSSIONS

$BaTiO_3$ is a ferroelectric material below 120 °C. At room temperature, the permittivity of this compound is nearly independent of frequency in the low-frequency region, although it decreases with frequency in the radiofrequency and higher-frequency regions [20]. Figure 1(a) represents the experimental data at room temperature. The dielectric constant of $BaTiO_3$ decreases so slowly that it can be considered a constant at the frequency range from 5 to 2500 Hz. The permittivity of silicone oil is also nearly a constant in the range of 5–2500 Hz; see Fig. 1(b). We suspended $BaTiO_3$ particles into silicone oil as ER fluid. When we adjust the frequency from 5 to 2500 Hz and hold the voltage amplitude U_0 on ER fluids, there is only one parameter, namely the frequency of the applied electric fields which influences the ER effect. Thus the system consisting of $BaTiO_3$ particles suspended in silicone oil should be an ideal system to investigate the frequency dependence of the ER effect.

In the ER fluids, the spherical particles are employed as the suspended solid particles, and a sinusoidal electric field $U = U_0 \sin(\omega t)$ is applied on ER fluids. Spherical particles and the sinusoidal wave will simplify the theoretical calculation of the ER effect.

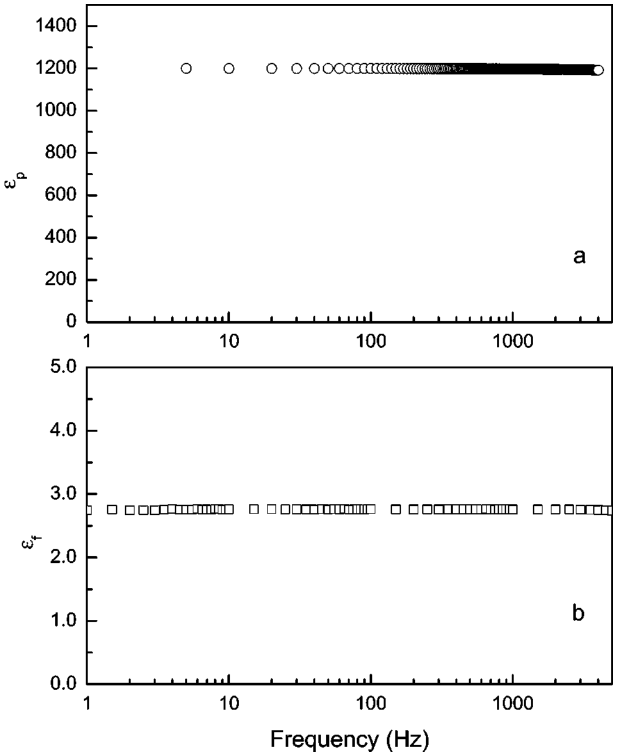

FIG. 1. Frequency dependence of dielectric constants of (a) $BaTiO_3$ and (b) silicone oil.

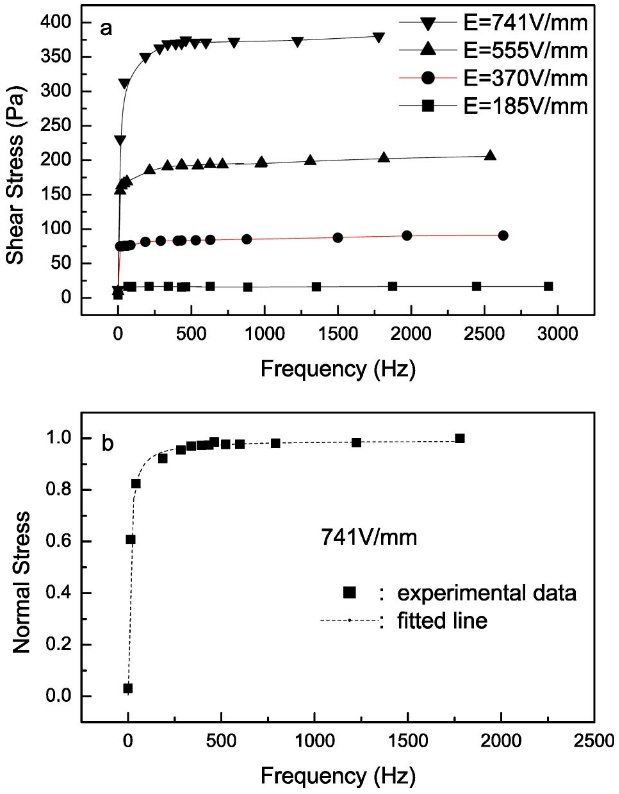

FIG. 2. (a) Shear stress of single-crystal $BaTiO_3$/silicone oil ER fluids at different electric fields; (b) normalized shear stress dependence of frequency at $E=741$ V/mm. Solid squares are experimental data. Dashed line is calculated according to Eq. (3). Volume ratio 32%, shear rate $\gamma=2.509$/s.

Both ac conductivity of the solid particles and that of the silicone oil increase with frequency linearly [19]. In our experimental range 5–2500 Hz, the conductivity follows $\sigma(\omega)=\sigma_0\omega$, where $\sigma_0(BaTiO_3)=1\times10^{-15}$ S/m, $\sigma_0(\text{silicone oil})=1.4\times10^{-13}$ S/m.

Fixing the effective voltage $|U|$ across ER fluids, the shear stress of ER fluids is measured at different frequency by adjusting the power supply. Figure 2(a) shows the shear stress versus frequency at different electric fields. Here the effective voltage $|U|=|U_0\sin(\omega t)|=(\sqrt{2}/2)U_0$ is used. The electric field $E=|U|/L$, where L is the gap between the bob and the cap of the rheometer. In the paper, the shear stress is defined as $\tau=\tau(E)-\tau(0)$, where $\tau(E)$ and $\tau(0)$ are the apparent shear stress of the ER fluids under electric field E and without electric field, respectively. τ is usually E^{γ} $(\gamma\sim1-2)$-dependent.

The experimental tendency is very similar to the theory proposed by Davis [21]. According to the theory, the shear stress increases sharply with frequency and saturates beyond 100 Hz. This is the first time to our knowledge, that a sharp increase with frequency is observed in experiments. In previous literature [5,9,10], the shear stress usually increases slowly with frequency in the range of 0–2500 Hz. If we calculate β^2 using Eq. (3) with parameters $\epsilon_p=1199$, $\epsilon_f=2.75$, $\sigma_p=4.03\times10^{-9}$ S/m, and $\sigma_f=7.47\times10^{-8}$ S/m, the theory fits the measured data very well [see the dashed line in Fig. 2(b)]. The excellent agreement indicates that the mismatch polarization model can explain the ER effect well in water-free and non-coating-particle ER systems. However, the fitted conductivities of the particles and the silicone oil are much higher than the measured values. The origin of the discrepancy can be explained in the field dependence of the conductivity.

In ER fluids, the local electric field between two particles, $E_{\text{local}}=E_0+(4.808/4\pi\varepsilon_0\varepsilon_f d^3)P$ (d is the distance between two particles), is usually much higher than the external applied electric fields [22]. Because the conductivity of $BaTiO_3$ varies with the strength of the electric fields [23] and the conductivity of the silicone oil is nonlinear [24], the higher local electric field will cause the much higher conductivities of $BaTiO_3$ particles and silicone oil. In other words, in calculation, we should use the conductivity under local electric fields (i.e., $\sigma_p=4.03\times10^{-9}$ S/m), not the experimental values measured at the low fields (i.e., $\sigma_p=1\times10^{-15}$ S/m).

Figure 3 shows the electrical current versus frequency. The current passing through ER fluids increases very slowly with frequency below 100 Hz, increases abruptly in the 100–500 Hz range (see the inset), and increases linearly with frequency in the high-frequency region, $I\propto\omega$. The Ohmic rule is only obeyed in the high-frequency region, not in the low-frequency region. The different behavior can be easily understood from Eq. (3). At low frequency, Eq. (3) can be reduced to $\beta_c\to(\sigma_p-\sigma_f)/(\sigma_p+2\sigma_f)$, and the conductivity mismatch dominates the ER effect. At high frequency, Eq. (3) is reduced to $\beta_d\to(\varepsilon_p-\varepsilon_f)/(\varepsilon_p+2\varepsilon_f)$, and the dielectric mismatch dominates the ER effect. With increasing frequency, the ER fluid passes from the conductivity-dominated regime to the dielectric regime. At a different region, the

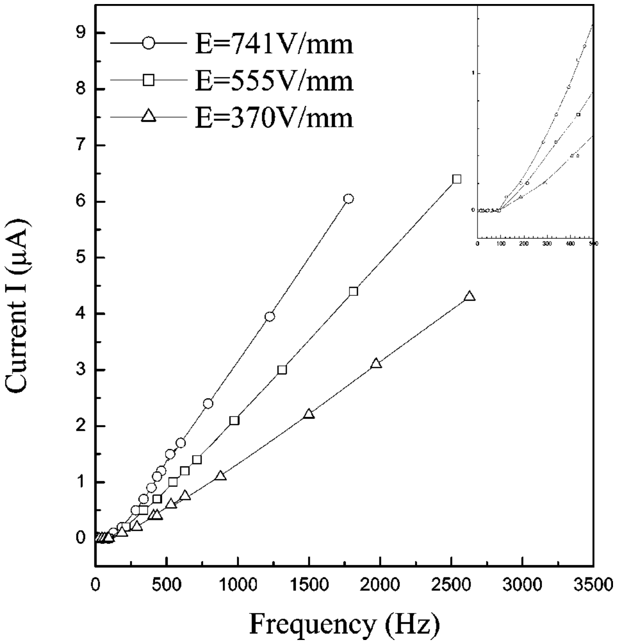

FIG. 3. Current vs frequency of single-crystal $BaTiO_3$/silicone oil ER fluids at different electric fields. Volume ratio 32%, shear rate $\gamma=2.509/\mathrm{s}$.

current shows different behavior with frequency. In the frequency region from 5 to 200 Hz, both the conductivity and the dielectric constant should play roles in the ER effect and the current shows a complex behavior. It is seems that 100 Hz is a *trans*-point from the conductivity-dominated regime to the dielectric regime.

The electric-field dependence of the shear stress at different frequency can give some clues to the polarization mechanism. Figure 4 gives the electric-field dependence of the shear stress for different volume fractions at different frequency. Under dc fields, the shear stress shows E dependence; see the inset in Fig. 4. At high frequency, the shear stress is E^2-dependent. It is obvious that the power of E approaches 2 with increasing frequency. It is also obvious that the ER effect shows different behavior at low frequency and high frequency, which indicates different mechanisms: the former is conductivity mismatch and the latter is dielectric mismatch.

Grinder *et al.* [25] investigated the time dependence of the barium titanate fluids with a unipolar square wave and a bipolar square wave at 5 and 10 Hz, respectively. The time dependence of the dipole moment was expressed as $P(t)=4\pi\varepsilon_0\varepsilon_f r^3 E_0[\beta_c+(\beta_d-\beta_c)e^{-t/\tau}]$, where the characteristic time $\tau=\varepsilon_0(\varepsilon_p+2\varepsilon_f)/(\sigma_p+2\sigma_f)$. They found the polarization is mainly dominated by the bulk polarization (conductivity mismatch and dielectric mismatch) while the surface polarization mechanism also exists. When polarized, the charge carries in the barium titanate (mainly holes) are carried to the particle surface on the time scale τ. It was concluded that the shear stress will increase with the frequency when frequency $f>1/\tau$ for the system (here $1/\tau\sim 30$ Hz). The conclusion supports our observation of the increasing of shear stress with frequency.

$BaTiO_3$ polycrystal spherical particles are also employed as ER fluid particles. Similar behavior has been observed; see Fig. 5. The shear stress also increases sharply with frequency below 100 Hz but increases slowly at high frequency over 250 Hz. No saturation is observed even up to 3000 Hz. The difference between single-crystal-particle ER fluids and polycrystal-particle ER fluids is probably due to the multidomain structure in polycrystal particles. However, more experimental study is needed to explain the phenomenon.

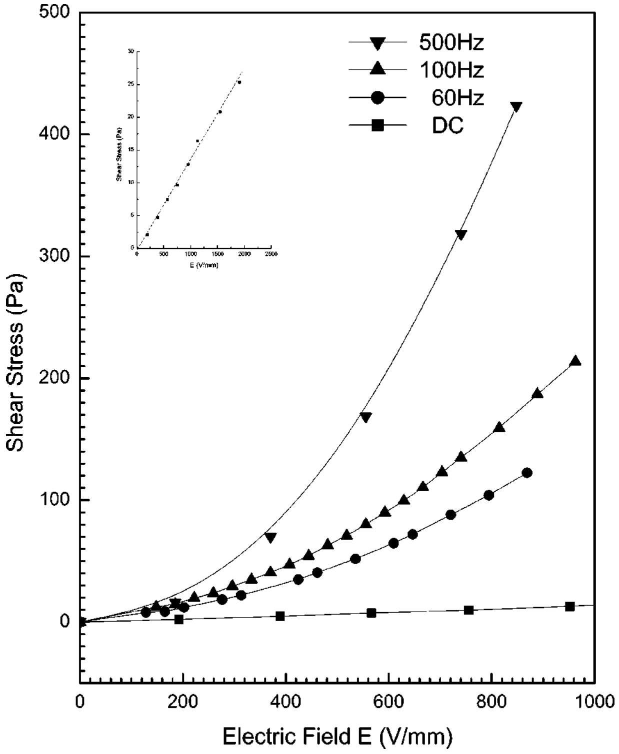

FIG. 4. Shear stress vs electric fields at different frequency in single-crytal $BaTiO_3$/silicone oil fluids. Inset is the enlargement of the shear stress dependence of electric fields under dc fields. Shear rate $\gamma=2.506/\mathrm{s}$, volume ratio 26%.

The frequency dependence of the shear stress for other systems, such as the triglycine sulfate [TGS $(CH_2CH_2COOH)H_2SO_4$]/silicone oil water-free system, has also been measured. Similar phenomena are observed. The shear stress also increases sharply with frequency below 100 Hz and saturates at high frequency. The dielectric constant of TGS particles is also independent of the frequency in the range of 5–2500 Hz [26].

All of the above experiments are carried out on water-free ER fluids. If water on the surface of the particles is not removed totally (for example, 0.06% water on the surface of the particles), the shear stress first decreases with frequency, down to 155 Pa at $f_c=86$ Hz, and increases until it saturates beyond 300 Hz, as shown in Fig. 6. The critical frequency f_c (at which the shear stress is lowest) increases with the size of the particles. If the water content is high (i.e., $>0.1\%$), the shear stress will decrease with frequency on the whole frequency range, as reported by Wen [27]. Ionic impurities on the interface of solid particles will redistribute and induce double-layer polarization under electric fields. Under these conditions, the interfacial polarization should be taken into account. In real ER fluids, the ER effect should be the sum of

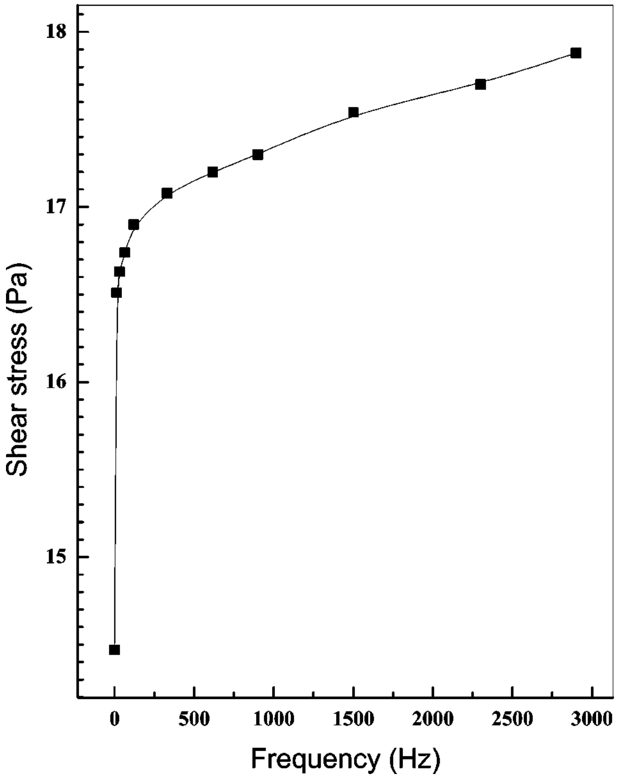

FIG. 5. Frequency dependence of the shear stress of polycrystal $BaTiO_3$ particles/silicone oil system. Volume ratio 10%, shear rate $\gamma=15.89/\mathrm{s}$, $E=365$ V/mm.

interfacial polarization and volume polarization [7,25]; the former usually decreases with frequency [2] while the latter increases [6,7]. Depending on which kind of polarization dominates the ER effect, the shear stress increases or decreases with the frequency of the external applied electric fields. When the water content on the particles is low or the surfactant is free, the volume polarization plays a role in the ER effect and the shear stress increases with frequency, as we observed here and reported in Ref. [5]. With the increase of water content, the interfacial polarization becomes more and more significant and dominates the ER effect, and the shear stress decreases with frequency, as in Ref. [27]. If the two polarizations are balanced, the shear stress is a constant with frequency or shows a complex relationship with frequency, as observed here. This should be the reason why we always observe a decrease of the ER effect with frequency in the surfactant-activated systems, while we observe an increase of the ER effect in many surfactant-free systems.

In addition, if the dielectric properties of the particles vary with frequency, the relationship between the shear stress and frequency will become more complex. In this case, the ER effect not only comes from the frequency, but also from the dielectric properties of particles, as shown in Ref. [6,7]. Most of the previous experiments have been carried out under this condition. However, because the permittivity of most used solid particles decreases greatly with frequency, as reported in [27,28], the frequency usually indirectly weakens the ER effect according to Eq. (3). Therefore, in many commercial ER fluids and in much of the reported literature, the ER effect always decreases with frequency.

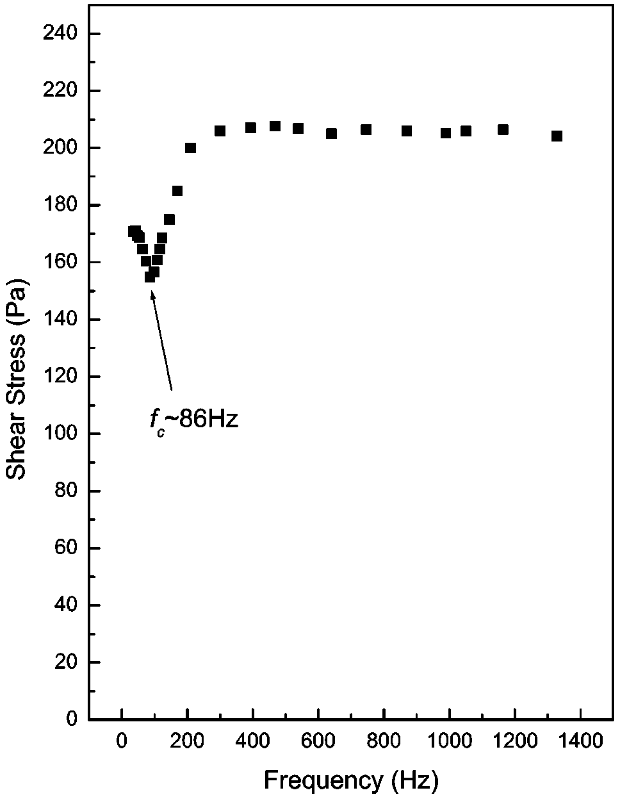

FIG. 6. Frequency dependence of shear stress of water-contained single-crystal $BaTiO_3$/silicone oil ER fluids. Water content is about 0.06%. Volume ratio 16%, $E=1000$ V/mm, shear rate $\gamma=2.509/\mathrm{s}$.

In summary, an ideal ER fluid system, $BaTiO_3$ single-crystal spheres/silicone oil, is employed to investigate the ER mechanism through measuring the frequency dependence of the ER effect. Because of the permittivity independence of the particles and the suspending liquid, the obtained experimental data reflect the reliable relationship between the ac frequency and the ER effect. In the experiments, spherical particles and sinusoidal ac electric fields are employed in order to simplify theoretical calculation. It is found that the shear stress increases sharply with frequency below 100 Hz and reaches saturated values quickly at high frequency. This phenomenon can be explained in terms of the mismatch of the permittivity between particles and the fluid. However, if there is some water in the systems, the interfacial polarization should be taken into account. The total ER effect should be the sum of two kinds of polarizations, namely interfacial polarization and volume polarization. Based on the experimental result, we give a reasonable explanation for the different ER-effect–frequency behaviors in the literature.

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