Structure-induced nonlinear dielectric properties in electrorheological fluids

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The structure-induced nonlinear dielectric properties of electrorheological (ER) fluids have been studied in anhydrous KNbO3–silicone oil and SrTiO3–silicone oil systems. We find that the permittivities of ER fluids increase linearly with the increase of the field strength as the electric field exceeds a threshold value $E_1$, and tend to saturate beyond a high field strength $E_2$. The alignment of particles in the direction of the electric field causes variation of the dielectric properties of the ER fluids. Below $E_1$, when the particles are randomly dispersed, and beyond $E_2$, when almost all the particles are form chains and columns, the permittivities do not vary with $E$ and attain their lowest and highest values, respectively. Correspondingly, the current densities of ER fluids no longer follow Ohm’s law between $E_1$ and $E_2$, whereas they still show Ohmic behavior in the regions below $E_1$ and beyond $E_2$. [S1063-651X(97)05302-6]

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I. INTRODUCTION

An electrorheological (ER) fluid consists of fine solid particles suspended in an insulating oil. The effective viscosity of an ER fluid increases dramatically and the particles form chains and columns when an electric field is applied and exceeds a critical value. These phenomena occur rapidly and are reversible. Much attention has been attracted to study the properties and the physical mechanism of ER fluids [1–6]. The basic issues are the field-induced interactions and behaviors of the particles in the fluids, which are related to the dielectric properties of these fluids. Although some studies of dielectric properties of ER fluids have been reported in literature, these mostly involve measurements under low electric field. However, the ER effect usually appears at high fields when rearrangement of the particles occurs. On the other hand, the treatment of the dielectric properties in conventional homogeneous two-phase mixture systems cannot be used here any longer, because the particles are movable and may aggregate in ER fluids. The most commonly noticed nonlinear electric property is that the current density increases more rapidly with the field strength $E$ than would be expected in Ohm behavior [6–8]. To date the explanation of this phenomenon is still insufficient. Block et al. [9] observed that the permittivity and dielectric loss were larger at high field than those at low field, and pointed out that the behavior was due to the fibrillation of particles in the field direction. Conrad et al. [10] found that the degree of alignment of the particles in the field direction increased with the field and then leveled off with further increase in the field. However, the relationship between the particle arrangement in the fluid and the nonlinear dielectric properties has not been established.

We have studied the nonlinear dielectric properties of some anhydrous ER fluids and found that the nonlinear behavior is mainly caused by the rearrangement of the particles in the fluids. The measured results show that the permittivities of ER fluids increase linearly with the increase of the field strength when the electric field exceeds a threshold value $E_1$, and then tend to saturated constants beyond a high field strength $E_2$. Below $E_1$ and above $E_2$, the permittivities are constant at their lowest and highest values, respectively. Correspondingly, the current density no longer follows Ohm’s law in the region between $E_1$ and $E_2$, while Ohm’s law is obeyed outside this region. A model based on the fact that the alignment of the particles in the electric field direction causes the permittivity to increase is proposed to explain the anomalous electric properties in ER fluids. The analysis is consistent with the measured results.

II. PERMITTIVITY AND CONDUCTIVITY OF A HETEROGENEOUS MIXTURE

The permittivity of a heterogeneous mixture has been studied by various authors [11]. In the case of spherical particles distributed in a medium, a useful formula for the permittivity was given by Looyenga [12]. The permittivity of an ER fluid under an electric field lower than the threshold value, in which the particles are randomly distributed, can be described approximately by Looyenga’s formula:

$$
epsilon_1 = [e_{f}^{1/3} + \phi (e_p^{1/3} - e_{f}^{1/3})]^{3},$$

where $e_p$ and $e_f$ are the permittivities of the particles and fluid, respectively, and $\phi$ is the volume fraction of the particles in the fluid.

Any aggregation of the particles along the direction of the electric field must lead to the increase of the permittivity of the system. This can be schematically illustrated as Fig. 1 and described as follows. For simplicity, we assume that all the particles dispersed in the oil are cubic. In Fig. 1(a), the particles are homogeneously distributed, and in Fig. 1(b), the particles are totally aligned in the electric-field direction. The permittivity of the system can be derived by regarding the admittance as the sum of all resistances and capacitances of solid and liquid blocks in series and in parallel, respectively.

The permittivity in the high frequency limit ($\omega \rightarrow \infty$) in the homogeneously distributed case is

$$
epsilon_1 = e_f \left( \frac{e_p + (\phi - \phi^{1/3})(e_p - e_f)}{e_p - \phi^{1/3}(e_p - e_f)} \right),$$

and in the aligned case is

$$
epsilon_1 = e_f \left( \frac{e_p + (\phi - \phi^{1/3})(e_p - e_f)}{e_p - \phi^{1/3}(e_p - e_f)} \right),$$

where $e_f$ is the permittivity of the fluid.
describing the behavior, although they are commonly ac-

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given in the Appendix, and a discussion will be made in a
to 

\( \varepsilon \), described by Eqs. (1) to (5), are plotted in Fig. 2.

In Fig. 2, curve (1) and curve (2) show the differences of the spherical particle model and the cubic particle model in the homogeneously distributed case, while curve (3) shows the permittivity and the conductivity of the system changing linearly with the volume fraction \( \phi \) in the aligned case. It can be seen that for a certain volume fraction \( \phi \) the permittivity \( \varepsilon \) or the conductivity \( \sigma \) of the system must increase when the particles are arranged in the electric-field direction.

\[ \varepsilon_{2, \infty} = \phi \varepsilon_p + (1 - \phi) \varepsilon_f. \]  
\( (3) \)

On the other hand, the conductivity \( \sigma \) in a dc field (\( \omega = 0 \)) in the homogeneously distributed case has an identical behavior with \( \varepsilon_{1, \infty} \), namely

\[ \sigma_1 = \sigma_f - \frac{\sigma_p + (\phi - \phi^{1/3})(\sigma_p - \sigma_f)}{\sigma_p - \phi^{1/3}(\sigma_p - \sigma_f)}. \]  
\( (4) \)

In the aligned case, the conductivity is

\[ \sigma_2 = \phi \sigma_p + (1 - \phi) \sigma_f. \]  
\( (5) \)

The derivations of Eqs. (2) to (5) are given in the Appendix. We should emphasize that a certain percentage of oil in the chains must contribute to the calculations in the aligned case, i.e., Eq. (3) and Eq. (5) are not appropriate for quantitatively describing the behavior, although they are commonly accepted in the literature [13]. More reliable representations are given in the Appendix, and a discussion will be made in a later part of this paper.

Our treatment with the cubic model is more intuitive. The behaviors of \( \varepsilon \) and \( \sigma \) changing with \( \phi \), described by Eqs. (1) to (5), are plotted in Fig. 2.

In Fig. 2, curve (1) and curve (2) show the differences of the spherical particle model and the cubic particle model in the homogeneously distributed case, while curve (3) shows the permittivity and the conductivity of the system changing linearly with the volume fraction \( \phi \) in the aligned case. It can be seen that for a certain volume fraction \( \phi \) the permittivity \( \varepsilon \) or the conductivity \( \sigma \) of the system must increase when the particles are arranged in the electric-field direction.

III. EXPERIMENTAL DETAILS

In the measurement of the permittivities of ER fluids, an ac high voltage power supply with sinusoidal voltage output was applied. Two parallel electrodes filled with an ER fluid constituted a capacitor in which the permittivity of the ER fluid was obtained by measuring the capacitances. The currents passing through the ER fluid were calculated by measuring the voltage on a resistance in series with the capacitor. The impedance of the resistance was three orders smaller than that of the capacitor. Special care was taken to avoid errors resulting from ac-circuit resonance effects. All digital multimeters and oscilloscopes applied in the experiments had high enough input impedances to ensure the reliability of all measured values.

Measurements were performed on two kinds of silicone oil ER fluids, one containing KNbO3 particles and the other SrTiO3 particles. Particles of average size 35 \( \mu \)m were prepared by grinding single crystals and drying above 200 °C in an oven to evaporate any water. At room temperature KNbO3 crystal is a ferroelectric material while SrTiO3 is not. The volume fractions of the particles for both KNbO3 and SrTiO3 in the oil were 0.05 and 0.2. The ER fluid container was installed in a silicone oil bath, the temperature of which could be controlled. Each value of the permittivity and the current density at different field strength was measured after the electric field had reached a fixed value for 20 s. All the current densities and field strengths were mean effective values measured under a 50-Hz ac field at room temperature.

IV. RESULTS AND DISCUSSIONS

The dependences of the current density and the permittivity of the KNbO3–silicone-oil ER fluid on an electric field are shown in Fig. 3(a) and Fig. 3(b), while those of the SrTiO3–silicone oil system are shown in Fig. 4(a) and Fig. 4(b), respectively. It is obvious that in a certain range of the electric field the permittivity increases linearly with the increasing field and the current density behaves nonlinearly in this region. In Fig. 3, the region is from about \( E_1 = 50 \) V/mm to \( E_2 = 2800 \) V/mm in the case of \( \phi = 0.2 \), while \( E_1 \) and \( E_2 \)
are lower at $\phi=0.05$. Below $E_1$, the permittivities are constants, and beyond $E_2$, they tend to saturated values. This fact is also explicit in the SrTiO$_3$–silicone oil system as shown in Fig. 4, where $E_1$ and $E_2$ are about 45 V/mm and 450 V/mm, respectively. In Fig. 3(a) and Fig. 4(a), the values of $E_1$ are not shown because they are hardly detectable.

![Figure 3](image1)

**FIG. 3.** Relation of (a) the current density and (b) the permittivity versus field strength in KNbO$_3$–silicone oil ER fluids ($\phi=0.05$ and 0.2). Inset shows the permittivity in the low field region. The solid lines across the symbols are guides to the eye.

The variation of permittivity is mainly due to the arrangement of the particles in the oil. When the field strength is lower than $E_1$, the particles in the fluid are randomly dispersed without rearrangement and the permittivity of the system does not change with the electric field, i.e., it is a constant $\varepsilon_1$. However, when the field strength is larger than $E_2$, all the particles almost form chains and columns in the direction of the field. Then the permittivity reaches $\varepsilon_2$ and maintains this value when the field strength is further increased. Equations (1) [or (2)] and (3), as well as Fig. 2, express the distinctness of the permittivities of these two extreme situations. In the region between $E_1$ and $E_2$, more and more particles are aligned along the direction of the field as the field strength increases, which causes the permittivity to increase. All the predictions have been demonstrated in our experiments through observation of the arrangement of the particles in ER fluids with an optical microscope. The observation was made simultaneously with the measurement of the permittivity by varying the electric field. Below threshold field $E_1$, no rearrangement of the particles could be observed. The aggregation of the particles in the direction of the field occurred at $E_1$, beyond which the particles were gradually aligned with the increase of the electric field. Finally, all particles formed chains or columns at $E_2$, above which the pattern was fixed and no more change could be observed.

Conrad et al. [10] measured the degree of alignment of the particles with an orientation parameter and found that the orientation parameter increases linearly with the electric field in a certain region of $E$ and tends to a constant for a larger field. It is reasonable to use Conrad’s results to explain the linear behavior of the permittivity increase with the electric field between $E_1$ and $E_2$ observed in our measurement. A further theoretical analysis about this phenomenon is still required.

These facts demonstrate that the polarization of the ER fluid between $E_1$ and $E_2$ is no longer linear, i.e., the ER fluid permittivity is $E$ dependent. The relation of the permittivity and electric field can be expressed as

$$\varepsilon = \begin{cases} \varepsilon_1, & E \leq E_1 \\ \varepsilon_1 + k(E-E_1), & E_1 < E < E_2 \\ \varepsilon_2 = \varepsilon_1 + k(E_2-E_1), & E \geq E_2 \end{cases} \quad (6)$$

where $k$ is a constant.

By the same analysis the conductivity of the ER fluid in a dc field is also $E$ dependent, i.e.,

$$\sigma = \begin{cases} \sigma_1, & E \leq E_1 \\ \sigma_1 + k'(E-E_1), & E_1 < E < E_2 \\ \sigma_2 = \sigma_1 + k'(E_2-E_1), & E \geq E_2 \end{cases} \quad (7)$$

where $k'$ is a constant different from $k$.

If we use the complex permittivity $\varepsilon_s(\varepsilon + j\varepsilon_0\omega)$ in Eq. (6), the effective permittivity should be $\varepsilon_s(\varepsilon^2 + \sigma^2/\varepsilon_0^2 \omega^2)^{1/2}$, where $\omega$ is the angular frequency. The magnitude of the current density in the ER fluid, which consists of displacement and conductance, can be written as

$$j = \omega \varepsilon_0 \left( \varepsilon^2 + \frac{\sigma^2}{\varepsilon_0^2 \omega^2} \right)^{1/2} E. \quad (8)$$
The current density in different regions corresponding to the permittivity and conductivity can then be expressed as follows. In the high frequency ac-field limit,

\[ j = \omega \varepsilon_0 \varepsilon E \]

\[ = \begin{cases} 
\omega \varepsilon_0 \varepsilon_1 E, & E \leq E_1 \\
\omega \varepsilon_0 \varepsilon_2 E = \omega \varepsilon_0 \varepsilon_1 + k(E - E_1), & E_1 < E < E_2 \\
\omega \varepsilon_0 E, & E \geq E_2 
\end{cases} \tag{9} \]

while in the case of a dc-field limit,

\[ j = \sigma E = \begin{cases} 
\sigma_1 E, & E \leq E_1 \\
\sigma_1 + k'(E - E_1), & E_1 < E < E_2 \\
\sigma_2 E, & E \geq E_2 
\end{cases} \tag{10} \]

Because of the nonlinear polarization or the nonlinear conductivity of the ER fluid in the region between \( E_1 \) and \( E_2 \), the \( E \) dependence of the current density behaves as \( j = aE + bE^2 \), where \( a = \omega \varepsilon_0 \varepsilon_1 - k \omega \varepsilon_0 E_1 \) and \( b = k \omega \varepsilon_0 \) in an ac field or \( a = \sigma_1 - k' E_1 \) and \( b = k' \) in a dc field.

A common expression for the current density can be obtained by combining Eqs. (6) to (8) and will be a little more complicated. It is obvious that the current density should also exhibit nonlinear behavior between \( E_1 \) and \( E_2 \).

In the regions below \( E_1 \) and above \( E_2 \), the current densities still follow Ohm’s law, but with different slopes. The slope in the region \( E \leq E_1 \) is usually much larger than that in \( E \leq E_1 \); in other words, the current density increases with \( E \) more steeply beyond \( E_2 \) than below \( E_1 \).

The field dependence of the current density \( j \) and permittivity \( \varepsilon \) or conductivity \( \sigma \) are schematically plotted in Figs. 5(a) and 5(b), respectively. With this analysis the measured behaviors of the permittivity and current density shown in Figs. 3 and 4 can be well understood. A quantitative comparison between calculation and measurement is difficult because of the arbitrary shape and the inhomogeneous distribution of the particles in the oil. In practice, even at \( E \geq E_2 \), not all chains and columns consisting of particles are fully filled in between electrodes, i.e., there are some chains and columns shorter than the distance between the two electrodes and there is more oil in these aligned blocks. However, in our model, by adjusting the parameter \( \chi \), which represents the average percentage of oil in each chain as described in the Appendix, we can calculate the permittivity to fit the measured value of the permittivity. For example, according to Eqs. (A3) and (A9) to (A12) in the Appendix, when the electric field strength is larger than \( E_2 \), the calculated permittivities of KNO\(_3\)-silicone-oil ER fluid for \( \phi=0.05 \) and 0.2 are 6.103 and 15.340, respectively, corresponding to two values of \( \chi \) (0.01 and 0.0145), which quantitatively agree with the experimental results: 6.17 and 15.48. However, if we let \( \chi=0 \), the calculated results would be 7.38 and 22.0, which are quite different from the measured values. This fact indicates that Eqs. (3) and (5) in the text or Eqs. (A14) and (A16) in the Appendix are not appropriate for describing the real case in which the particles are not cubic and not all the chains or columns are spanning the two electrodes. A better approximation should be Eqs. (A9) to (A12) in the Appendix. However, when the field strength is smaller than \( E_1 \), the calculated results do not fit so well as those at \( E \geq E_2 \). The main reason is that the particles cannot be homogeneously distributed in the oil, and their shapes are arbitrary. Nevertheless, the model we have described can explain the reason why the permittivity and the conductivity maintain linearity when \( E \leq E_1 \) and \( E \geq E_2 \) and should behave nonlinearly when \( E_1 < E < E_2 \).

In our experiment, because ER fluids consist of anhydrous particles and silicone oil, the relaxation is not important in the case when the field frequency is not very high. The Looyenga equation and the model we have proposed are valid. However, if ER fluids contain water or mobile ions, the relaxation problem may be serious and should be taken into account to study the nonlinear behavior. Although other factors may also contribute to the nonlinearity of the dielectric property, the structure-induced nonlinear behavior is always most important in ER fluids.

V. CONCLUSIONS

In this article the structure-induced nonlinear dielectric properties of anhydrous ER fluids have been studied. The field dependence of the permittivities and the current densities of KNO\(_3\)-silicone oil system as well as SrTiO\(_3\)-silicone oil ER fluids were measured. The nonlinear behavior is mainly caused by the arrangement of the particles in the fluids. We have found that the permittivities of the ER fluids increase linearly with the increase of the field strength in the region between a threshold field \( E_1 \) and a critical field \( E_2 \). The enhancement of the permittivities is induced by gradual alignment of particles in the direction of the electric field. Below \( E_1 \), when the particles are randomly dispersed, and beyond \( E_2 \), when the particles almost totally form chains and columns, the permittivities do not vary with \( E \) and reach their lowest and highest values, respectively. Correspondingly, the current densities of the ER fluids behave as \( j = aE + bE^2 \) between \( E_1 \) and \( E_2 \), whereas they still follow Ohm’s law in the regions below \( E_1 \) and beyond \( E_2 \). This analysis about the structure-induced nonlinear behavior is valid in anhydrous ER fluids and can also be applied in other...
Assuming that \( N \) cubic particles of side length \( a \) are randomly dispersed in the oil, the ER fluid is divided into \( N \) cubes with side length \( a_f \), and each cube encloses a cubic particle; \( d \) is the distance between the two parallel electrodes of which the surface areas are both \( S \) [Fig. 6(a)]. The total volume of the ER fluid and the solid particles are \( V = Na_f^3 \) and \( V_p = Na^3 \), respectively. The volume fraction is \( \phi = V_p/V \), and \( a_f = \phi^{-1/3}a \).

The total admittance \( Y_s \) of the ER fluid can be represented as a resistance \( R \) and a capacitance \( C \) in parallel, i.e.,

\[
Y_s = \frac{1}{R} + i\omega C = \sigma \frac{S}{d} + i\omega\varepsilon_0(e' - i e'') \frac{S}{d}, \tag{A1}
\]

where \( \sigma, e', \) and \( e'' \) are the dc conductivity, and the real and imaginary parts of the dielectric constant of the fluid, respectively. According to Fig. 6(a), the cubes are in series in the field direction, but in parallel in the plane vertical to the direction of the electric field. Assuming that both components are loss-free, and their permittivities keep constant in the frequency region considered, then the total admittance \( Y_s \) can be rewritten

\[
Y_s = \frac{S}{d} \left[ \sigma + \omega \varepsilon_0 \left( e_x + \frac{e_x - e_{xx}}{1 + \omega^2 \tau^2} \right) \right]. \tag{A2}
\]

Comparing Eq. (A2) with Eq. (A1) leads to the Debye equation [11]:

\[
e' = e_x + \frac{e_x - e_{xx}}{1 + \omega^2 \tau^2}, \tag{A3}
\]

\[
e'' = \frac{(e_x - e_{xx}) \omega \tau}{1 + \omega^2 \tau^2}, \tag{A4}
\]

where

\[
\begin{align*}
\varepsilon_p &= \frac{\sigma_p^2 e_f - \phi \frac{1}{13} (\sigma_p^2 e_f - \sigma_f^2 e_p)}{[\sigma_p - \phi \frac{1}{13} (\sigma_p - \sigma_f)]^2} + (\phi - \phi^{1/3}) \\
&\quad \times \frac{(\sigma_p^2 e_f + \sigma_f^2 e_p - 2 \sigma_p \sigma_f e_f - \phi \frac{1}{13} e_f (\sigma_p - \sigma_f)^2)}{[\sigma_p - \phi \frac{1}{13} (\sigma_p - \sigma_f)]^2}, \tag{A5}
\end{align*}
\]

\[
\begin{align*}
e_x &= e_f \frac{e_p + (\phi - \phi^{1/3})(e_p - e_f)}{e_p - \phi^{1/3}(e_p - e_f)}, \tag{A6}
\end{align*}
\]

\[
\begin{align*}
\sigma &= \sigma_f + (\phi - \phi^{1/3})(\sigma_p - \sigma_f) \quad \frac{1}{\sigma_p - \phi^{1/3}(\sigma_p - \sigma_f)}, \tag{A7}
\end{align*}
\]

and

\[
\tau = \frac{e_p - \phi^{1/3}(e_p - e_f)}{e_p - \phi^{1/3}(\sigma_p - \sigma_f)}. \tag{A8}
\]

In the above equations, \( e_i \) and \( e_{xx} \) are the static and high-frequency dielectric constant of the ER fluid, \( \tau \) is the relaxation time, while \( \sigma_f, e_f, \) and \( \sigma_p, e_p \) are the conductivity and permittivity of the particles and the oil, respectively.

In fact, not all particles are properly enclosed by the corresponding oil cubes with side length \( a_f \). In order to improve the precision of the model, we discard the notation of oil cubes and each cube encloses a cubic particle, and each cube encloses a cubic particle. Assuming that both cubes are aligned in the direction of the electric field with a thin layer of oil between neighboring particles, although more factors should be considered.

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APPENDIX

In Fig. 6(b), the solid particles are assumed to be randomly distributed in a medium between two electrodes. (a) Randomly distributed, (b) aligned in the direction of the electric field with a thin layer of oil between neighboring particles.
Assuming that each chain is completely filled with particles, i.e., \( x \to 0 \) [Fig. 1(b)], then Eqs. (A9) to (A12) become

\[
\varepsilon' = \varepsilon'' = \phi \varepsilon_p + (1 - \phi) \varepsilon_f, \tag{A13}
\]

and

\[
\sigma' = \phi \sigma_p + (1 - \phi) \sigma_f, \tag{A14}
\]

and Eqs. (3) and (4) also transform into

\[
\varepsilon' = \varepsilon'' = \phi \varepsilon_p + (1 - \phi) \varepsilon_f, \tag{A16}
\]

and

\[
\varepsilon'' = 0. \tag{A17}
\]

With the increase of the field strength, the particles aggregate along the field direction gradually. Thus, the number of the particles in one chain increases, whereas the chain number and the total thickness of the oil layers thus the parameter \( x \) in one chain decreases. As the electric field exceeds the critical value \( E_2 \), the structure reaches its steady state and no longer changes with the external electric field.