

## Orientation of particles in an electrorheological fluid under an electric field

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The orientation of the particles in an electrorheological fluid under an electric field has been studied. Triglycine sulfate (TGS)–wax mixtures were applied in this study. Here the TGS particles are in the ferroelectric phase with spontaneous polarization  $\mathbf{p}_s$  in the  $\langle 010 \rangle$  direction at below the Curie temperature  $T_C$  and become paraelectric without a permanent electric moment at above  $T_C$ . By measuring x-ray-diffraction intensities of TGS/wax samples under an electric field at below and above  $T_C$ , respectively, it is found that the particles with spontaneous polarization turn their  $\mathbf{p}_s$  vectors to the direction of the external electric field in addition to forming chains or columns. However, the particles without permanent moment only form chains or columns, in which no preferred orientation of the particles can be observed. The preferred orientation degree is determined by comparing the diffraction intensity in these two cases. [S1063-651X(99)03110-4]

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### I. INTRODUCTION

An electrorheological (ER) fluid, consisting of solid particles and insulating liquid, can be transformed into a solid-like state when an electric field is applied. According to the polarization theory, the induced polarization of the particles  $\mathbf{p}_{\text{ind}}$  is dependent on  $\epsilon_p$ ,  $\epsilon_f$  (the permittivity of the particles and liquid, respectively), and  $R$  (the radius of the particles) and can be presented as  $p_{\text{ind}} = 4\pi\epsilon_0\epsilon_f R^3 E(\epsilon_p - \epsilon_f)/(\epsilon_p + 2\epsilon_f)$ . The interaction of the induced polarizations  $\mathbf{p}_{\text{ind}}$  leads the particles to form chains or columns and causes the increase of the shear stress in the ER fluids [1–6]. However, other certain parameters appear to have been overlooked because there is as yet no clear explanation for the quantitative difference between the experimental data and theoretical calculations [7,8]. The significantly different frequency responses, the shear stress increases with the frequency of electric field [9], or the decreases with the frequency [10], for instance, indicate that besides the structural change under an electric field, some other factors may play a role in the ER effect. Sometimes the ferroelectric particles are used for preparing the ER fluid, in which those particles with a spontaneous polarization moment must be affected by the external electric field. It is easy to imagine that the particles with a permanent dipole moment should receive the torques to orient them to the direction of the electric field.

A previous report revealed the turning of ferroelectric microspheres contained in ER fluids under a dc electric field [11]. However, the study of the effect was still incomplete. In the present paper, we have measured the orientation of the particles with a permanent dipole moment, which is compared with the case of particles without a permanent moment, in the ER fluid under an electric field. In order to obtain a reliable result for a quantitative comparison, the particles, which belong to the ferroelectric phase with spontaneous polarization moment at below Curie temperature  $T_C$  and become the paraelectric phase without a permanent moment at above  $T_C$ , were applied in the experiment. The

chemical nature, volume fraction, as well as the size and shape of the particles are all the same in the prepared ER fluids. The only different character is with and without spontaneous polarization at below and above  $T_C$ . The orientation degree of the particles in the suspensions must depend on this character under an electric field. With the x-ray-diffraction measurements, it is found that the vectors of the permanent dipole moment of most particles point in the direction of an external field, while no orientation can be observed in the case of the particles without permanent dipole moment, when an electric field is applied. In addition, the degree of the preferred orientation of the particles is quantitatively determined and a method to calculate the average dielectric property of the particles in the suspension is proposed in the ER fluids containing the ferroelectric particles.

### II. EXPERIMENT

TGS [triglycine sulfate  $(\text{NH}_3\text{CH}_2\text{COOH})_3 \cdot \text{H}_2\text{SO}_4$ ], a ferroelectric material, was applied for preparing the ER fluid, of which the Curie temperature is  $50^\circ\text{C}$ . TGS has a spontaneous polarization moment  $\mathbf{p}_s$  in the  $\langle 010 \rangle$  direction when the temperature is lower than  $50^\circ\text{C}$  and belongs to the paraelectric phase without spontaneous polarization at above  $50^\circ\text{C}$  [12]. The particles, obtained by grinding single-crystal TGS into powder with an average diameter of  $30\ \mu\text{m}$ , were suspended in the waxes. Two kinds of waxes, i.e., wax 1 and wax 2, of which the melting points were  $46$  and  $58^\circ\text{C}$ , were used as matrixes for mixing the TGS particles with a volume fraction of 10%, respectively. Three samples were prepared: two TGS/wax 1 mixtures and one TGS/wax 2 mixture. The TGS/wax 1 mixtures were heated to the temperature  $T_{\text{prep}} = 48^\circ\text{C}$  and TGS/wax 2 mixture to  $T_{\text{prep}} = 60^\circ\text{C}$ , respectively, where the waxes were melted to the liquids. Then the mixtures were stirred to uniform suspensions at  $T_{\text{prep}}$ . One (sample A) of the TGS/wax 1 samples was cooled down to room temperature in the absence of an electric field, becoming a solid mixture with the random distribution of TGS particles in it. Another TGS/wax 1 (sample B) and TGS/wax 2 (sample C) were applied to an ac field of  $1500\ \text{V/mm}$  at  $48$  and  $60^\circ\text{C}$ , respectively. The chains or columns of the TGS

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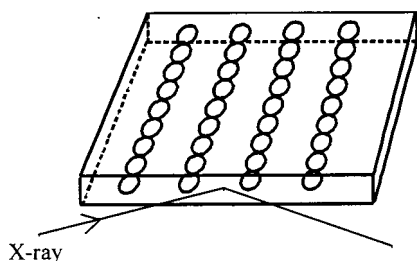


FIG. 1. Schematic of the sample A, B, and C in x-ray-diffraction measurements.

particles in sample B and sample C were induced by the applied electric field as in the usual ER fluids, which were observed with an optical microscope. When the chains or columns were stabilized, the suspensions were cooled down to room temperature with the electric field applied. After the suspensions were frozen, the electric field was removed. All three solid samples were cut into pieces for x-ray-diffraction measurements at room temperature.

The x-ray-diffraction (XRD) measurements were carried out with a Cu  $K\alpha$  radiation at room temperature. The voltage and the current were fixed at 50 kV and 200 mA, respectively. The schematic x-ray incidence on the samples is shown in Fig. 1.

### III. RESULTS AND DISCUSSION

From the microscopic observation, the chain or column structures of the particles formed along the electric field in the frozen samples B and C were the same as that at  $T_{\text{prep}}$ , while there was a random distribution for the particles in sample A because of no electric field applied.

The x-ray-diffraction spectra of samples A, B, and C are shown in Figs. 2A, 2B, and 2C, respectively, in which the diffraction peaks of the solid waxes are subtracted.

Figure 2A is the diffraction pattern of sample A quenched from 48 °C with no electric field applied. Although the TGS particles are in the ferroelectric phase with spontaneous polarization  $\mathbf{p}_s$ , the polarization can point in any direction in the absence of an external field. Therefore, the particles are randomly distributed without any preferred orientations as shown in Fig. 3(a). It can be regarded as a normal powder-diffraction pattern.

In the case of sample C, TGS has no permanent dipole moment ( $\mathbf{p}_s = \mathbf{0}$ ) at  $T > 50$  °C. Thus, the particles interact with each other only by the induced dipole moment  $\mathbf{p}_{\text{ind}}$  under an electric field and form chains or columns, but the  $\langle 010 \rangle$  of particles is distributed randomly, as shown in Fig. 3(c). When the sample is cooled through the frozen point 58 °C of wax 2 down to room temperature, all of the particles have been fixed in the wax. Although the TGS particles belong to the ferroelectric phase with spontaneous polarization moment  $\mathbf{p}_s$  in the temperatures below 50 °C, those particles cannot be turned any more in the solid wax by the effect of external electric field. It can be seen from Fig. 2C that no obvious preferred orientation appears in the diffraction pattern of the sample C, which is almost the same as that of sample A.

The sample B was applied at an electric field at 48 °C

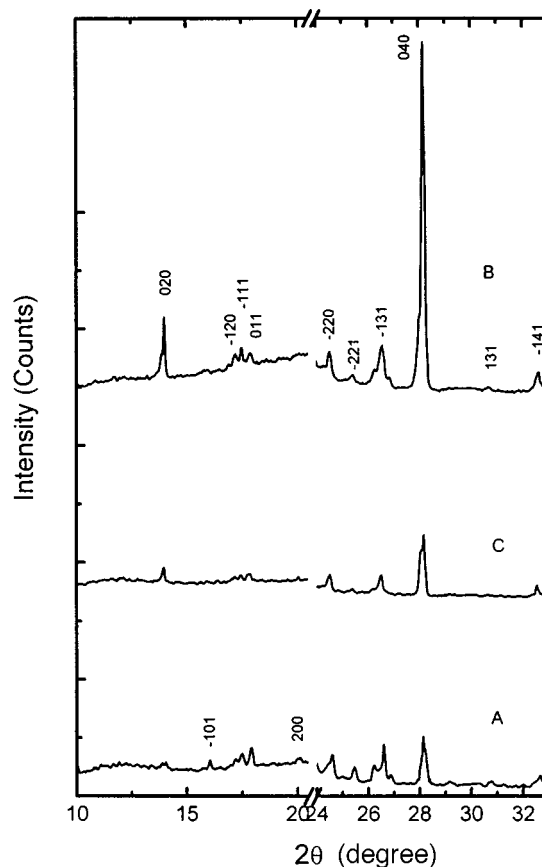


FIG. 2. The x-ray-diffraction spectra of sample A, B, and C. A, sample A,  $E = 0$  V/mm; B, sample B,  $E = 1500$  V/mm; C, sample C,  $E = 1500$  V/mm.

below the Curie temperature. The TGS particles possess spontaneous polarization  $\mathbf{p}_s$ , which must be affected by the external field. Therefore, in the case of sample B, it can be seen from Fig. 2B that the intensities of the (020) and (040) diffraction peaks increase dramatically while that of (200) is reduced compared to that of sample A and C. This is due to the preferred orientation of the particles with  $\mathbf{p}_s$  in the direction of the electric field. In the electric field, the induced polarization  $\mathbf{p}_{\text{ind}}$  leads the TGS particles forming chains or columns along the electric field, while the spontaneous polarizations  $\mathbf{p}_s$  cause the particles to receive a torque  $\Gamma = p_s E$  and turn towards the direction of the field. Because  $\mathbf{p}_s$  of TGS is parallel to  $\langle 010 \rangle$ , the particles will rotate to make (0k0) planes, i.e., (020), (040), etc., perpendicular to the direction of the electric field. Therefore, the intensities of the (020) and (040) peaks will greatly increase and the intensity of (200) diffraction decreases.

The relative intensity ratio of the diffraction peaks between sample B and sample A is plotted in Fig. 4.  $I_B$  represents the x-ray-diffraction intensities of sample B under an electric field of 1500 V/mm and  $I_A$  is that of sample A without electric field applied. It is obvious that the amplitudes of (020) and (040) increase dramatically when an electric field is applied, i.e., the preferred orientation of particles is in the direction of  $\langle 010 \rangle$  along the electric field.

A quantitative calculation of the preferring orientation degree is given as follows. The preferred orientation degree  $F$  is generally defined as  $F = (P - P_0)/(1 - P_0)$ , where  $P$  is the

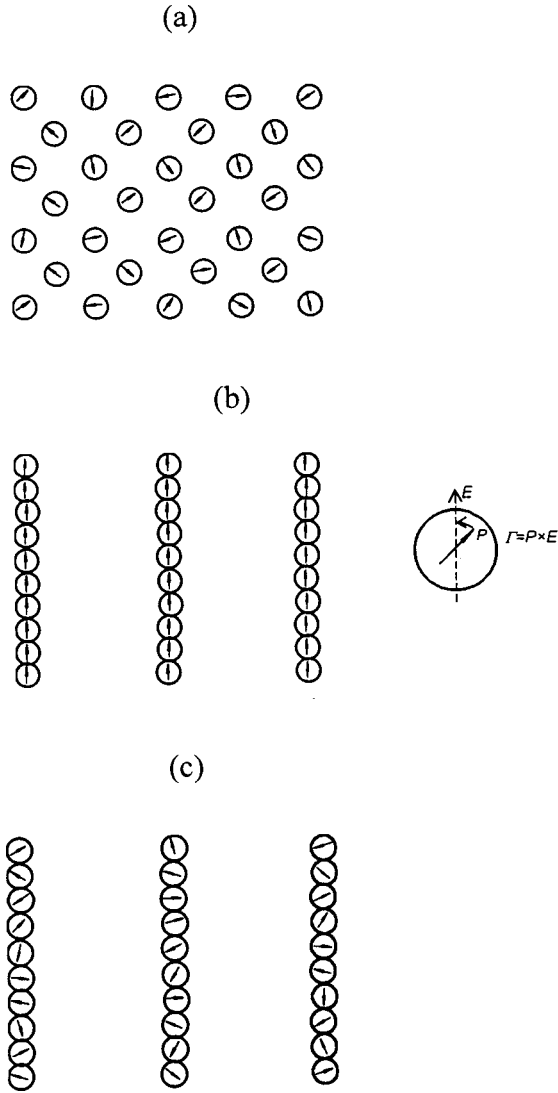


FIG. 3. Schematic representation of the particle distribution. The arrows represent the direction of  $\langle 010 \rangle$ . (a) In sample A, the particles randomly distributed;  $\mathbf{p}_{\text{ind}}=0$  and  $\mathbf{p}_s$  in the  $\langle 010 \rangle$  direction. (b) In sample B, the particles with preferring orientation; both  $\mathbf{p}_{\text{ind}}$  and  $\mathbf{p}_s$  are in the direction of the electric field. (c) In sample C, the particles without preferring orientation;  $\mathbf{p}_{\text{ind}}$  in the direction of the field and  $\mathbf{p}_s=0$ .

ratio of the sum of  $(0k0)$  diffraction intensities to the sum of all  $(hkl)$  diffraction intensities in the samples with orientated particles (for instance, in sample B), and  $P_0$  is that without orientation (for instance, in sample A). If  $F=1$ , the  $\langle 010 \rangle$  of all TGS particles are parallel to the direction of electric field. If  $F=0$ , the particles are randomly distributed. The average  $F$  in sample B was calculated by using the data withdrawn from Figs. 2B and 2A. Then the preferred orientation degree  $F=76\%$  is obtained. In the calculation of  $F$ , a simplified formula [13] was applied.

The fact that the preferred orientation degree  $F=76\%$  in sample B is less than the ideal value 100% indicates that the particles with the permanent moment  $\mathbf{p}_s$  in TGS/wax mixture are mostly orientated to the direction of the electric field. The deviation in the orientations of the particles may be due to the polydomain contained in some ferroelectric TGS particles or the disturbance of the interaction from the induced polarization of the particles.

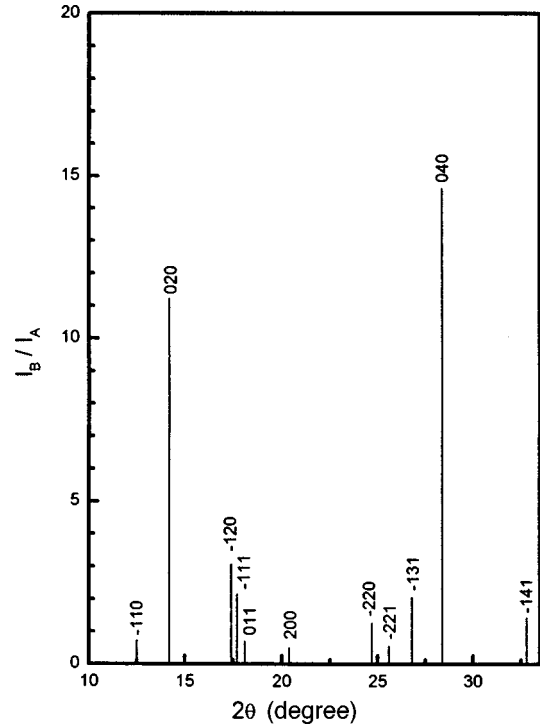


FIG. 4. The ratio of the diffraction intensities between sample B and sample A.

Based on the data of the preferred orientation degree, an equation can be proposed to calculate the average complex permittivity of anisotropic particles in the mixture:

$$\varepsilon_p^* = F \varepsilon_{p,b}^* + (1-F) \frac{\varepsilon_{p,a}^* + \varepsilon_{p,b}^* + \varepsilon_{p,c}^*}{3},$$

where  $a, b, c$  refer to three crystal axes of the particles,  $b$  is the polarization axis, and  $\varepsilon_p^*$  is the complex dielectric constant of the particles. By considering the orientation of the particles in calculating mismatch factor  $\beta^* = (\varepsilon_p^* - \varepsilon_f^*) / (\varepsilon_p^* + 2\varepsilon_f^*)$ , a better result was achieved for considering the properties in ER fluids [7,8].

#### IV. CONCLUSIONS

We have studied the orientation of the particles in ER fluid under an electric field. TGS/wax mixtures, in which the TGS particles belong to the ferroelectric phase with spontaneous polarization  $\mathbf{p}_s$  at below the Curie temperature  $T_C$  and become the paraelectric phase without permanent electric moment at above  $T_C$ , were applied in the study. By measuring x-ray-diffraction intensities of TGS/wax samples under an electric field at below and above  $T_C$ , respectively, it is found that the particles with  $\mathbf{p}_s$  receive torques and mostly turn their  $\mathbf{p}_s$  vectors to the direction of the external electric field. However, the particles without permanent moment only form chains or columns, in which no preferred orientation of the particles can be observed. The preferred orientation degree is determined by comparing the diffraction intensity changes in these two cases. The related properties of the suspensions consisting of anisotropic particles must be af-

fectured by the orientation of the particles in an electric field. This approach can also be used to study other suspensions containing ferroelectric particles under electric fields or containing ferromagnetic particles under magnetic fields.

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