Electrical field-driven ripening profiles of colloidal suspensions*

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Electrorheological (ER) fluid is a type of smart fluid whose shear yield stress relies on the external electrical field strength. The transition of ER fluid microstructure driven by the electrical field is the reason why viscosity changes. Experimentally, the transparent electrodes are used to investigate the column size distribution where an external electric field is applied to a colloidal suspension, i.e., ER fluid is increased. The coarsening profile of ER suspensions is strongly related to electrical field strength, but it is insensitive to particle size. In addition, in a low field range the shear stress corresponding to the mean column diameter is studied and they are found to satisfy a power law. However, this dependence is invalid when the field strength surpasses a threshold value.

Keywords: electrorheological fluid, electrical field, coarsening structure

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1. Introduction

The electrorheological (ER) fluid, a kind of typical colloidal suspension, is usually composed of high dielectric constant solid particles uniformly dispersed in a low dielectric constant insulating oil. The common ER fluid's dispersants are silicone oil and mineral oil. Since ER fluid was discovered, researchers have tried to search for a variety of materials as ER fluid's dispersed phases, for example, aluminum powder, starch, silicon, titanium dioxide, semiconductors, etc. The properties of dispersed phases determine the performance of the ER effect.[1] Wen *et al.* used nanoparticles ($BaTiO(C_2O_4)_2 \cdot NH_2CONH_2$) coated with urea as the dispersed phase and thus created the first kind of giant electrorheological (GER) fluid. The solid state can reach a yield stress of 130 kPa, which breaks the theoretical upper bound on conventional ER static yield stress.[2] Lu *et al.* reported that the polar molecules dominated the electrorheological (PM-ER) effect and developed a new PM-ER fluid whose yield stress can reach the order of MPa.[3] The ER effect is reversibly changed in the apparent viscosity of a colloidal fluid under an external electric field. A typical ER fluid consists of solid dielectric particles dispersed in an insulating fluid. It is generally believed that the large increase in the viscosity of ER fluid is caused by field-induced dipole–dipole interactions among the particles.[1,4,8] Previous studies have shown that the rheological response of ER fluid is related to structural changes when the electrical field strength is increased.[9,10]

Many studies have been conducted on the structural formation of ER fluid and computer simulation on the structural formation has been performed.[11] The relationship between the ER fluid microscopic structure and its mechanical and optical properties has aroused considerable interest in the last decade, particularly in relation to the magnetorheological (MR) fluid. The reflection coefficients of TiO_3-coated Kaolinite and Y-doped $BaTiO_3$ based ER fluids increased with electrical field strength increasing, which is due to the change of ER fluid structure.[12] Theoretical computation has provided the basis for modeling the analysis of structural evolutions under rapidly increasing external magnetic fields. Results have shown that columns and wall-like structures are formed parallelly to the field direction.[13,14] Wu *et al.* studied the influence of the particle size on the rheological property of magnetic fluid both in experiment and in computer simulation.[15] Further work has investigated the pattern formation of these structures with the effect of the system particle volume fraction.[16] The dynamic response of the MR fluid to rotating magnetic fields has been a hot research area. Experiments revealed that the magnetic particles preferred to form isotropic clusters and isolated particles appeared, which are governed by the Mason number (the ratio of viscosity to magnetic force).[17] The time-dependent cluster–cluster aggregations of superparamagnetic particles were also analyzed to investigate the characteristics of cluster dynamics.[18] Similar studies have been conducted on the ER fluid. Kittipoomwong *et al.* investigated the evolutions of both the rheological properties and the suspension structure, and the transient response of electrorheological suspensions in shear flow subjected to a suddenly imposed electric field experimentally.[19] The response of shear stress of ER suspensions to the electric field has been studied theoretically.[20] The kinetics of structural formation in ER suspensions has been investigated using microchannel and high-speed microscopic imaging, showing that the nonuni-

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form growth of the particle structures in the channel was correlated to field strength and flow rate.[21]

Wen *et al.* experimentally observed the dynamic process of chain and column formations as a function of time under the influence of a high-speed switch power supply.[22,23] Wen *et al.* found that once the time of field strength applied to ER fluid surpasses a certain time length, the particles first form chains, and these chains then coarsen and eventually form columns spanning between two electrodes. Although we have observed the chain and column profiles earlier and analyzed the structures, mechanisms and dynamics of ER fluid,[24,25] a direct observation of the size distribution of column diameters has not been reported to date.

In this work, an experimental approach to using transparent electrodes to study the column distribution issue is reported. We present the experimental evidence of the particle aggregating processes as a function of field strength. Our findings in the present study are as follows. The size distributions of columns are found to be dependent on the field strength applied to the ER fluid, but independent of the particle size. Under a low electrical field strength, the interaction among particles increases exponentially as a function of the column diameter, which does not occur under high field strength. The column diameter is saturated when the field strength surpasses a critical value. The interaction among particles, nevertheless, is not saturated even though the column size remains unchanged. At a fixed field strength, the columns are very stable and no obvious change is observed as time passes by.

2. Experiments

Two kinds of ER fluid cells were used to observe the process of column formation. For a top view of the column shape, ER fluid containing the particle/liquid mixture was sandwiched between two indium–tin oxide (ITO) glasses. The two transparent electrodes with a spacing of 2 mm were connected to a high voltage power supply. For the side view, two parallel brass electrodes were mounted on a glass slide to form a cell with a volume of about 125 mm^3 (2.5 mm×10 mm×5 mm). An optical microscope with a video recorder was used to monitor the process of column formation as the field strength was increased. The images were taken after the columns had been stabilized. Glass microspheres respectively with diameters of 0.5 μm and 47 μm are used in this work. These solid particles were dispersed in silicone oil with a volume fraction of 0.15.±0.02. The strength of the shear stress of the ER fluid versus field strength was measured by a rotational viscometer at a fixed shear rate (shear rate: 12 s^{-1}). A rotating inner bob and a stationary outer cup were used with a high power supply. The spacing between the bob and cup was 1.5 mm. All experiments were performed at room temperature.

3. Results and discussion

The column formation from the top and the side with 0.5-μm glass microspheres dispersed in silicone oil are shown in Fig. 1. Figure 1(a) shows that at zero field, the particles are suspended in the oil randomly and Brownian motion cannot be ignored due to the small particle size. When the electrical field strength was increased to 300 V/mm, the particles began to aggregate to form thin columns with an average diameter of more than 100 μm. The light could transmit from the bottom to the top through two ITO transparent electrodes, making it easy to identify the column distribution [see Fig. 1(b)]. When the field strength is increased, the columns began to coarsen, which can be seen in Fig. 1(c). In the process of column coarsening, columns are combined gradually. As a result, with electrical field strength increasing, the number of columns in the unit area decreases and the proportion of the column section area first increases and then decreases as shown in Table 1. Figure 1(d) shows the final state of the ER fluid when the field strength surpasses 1500 V/mm. After that, no obvious variation in column diameter can be observed even though the field strength is further increased. Figures 1(e) and 1(f) show the side views of the ER structures respectively corresponding to Figs. 1(b) and 1(d). Here the columns, shown from the top view in Fig. 1(d), are connected to each other on the ITO surface. The columns are therefore not clearly identified as shown in the side view in Fig. 1(f).

Table 1. Statistical data of Figs. 1(g)–1(i).

Panels in Fig. 1	Number of columns	Proportion of column section area in the figures
(g)	71	27.288%
(h)	29	35.088%
(i)	24	29.730%

To investigate the size distribution profiles of columns at different electrical field strengths, the number of columns is counted and the diameter of each column is measured. It is very difficult to measure the column diameter, due to the irregular shapes of the columns and because the clusters are connected as observed from the top view. In our measurements the diameter of each column is obtained by averaging the sizes measured along the longest and shortest axes of column. More precisely, as the column has clear contrast to the background, we use software (Photoshop, Adobe; ImageJ, National Institutes of Health; NIS-Element, Nikon) to analyse the pictures thus to distinguish the columns and compute the volumes. The software could outline the profiles and columns and count the number. The diameter of the largest circumscribed circle of column profile is defined as the diameter of this column, which is shown in Figs. 1(g)–1(i) and Figs. 4(b)–4(d).

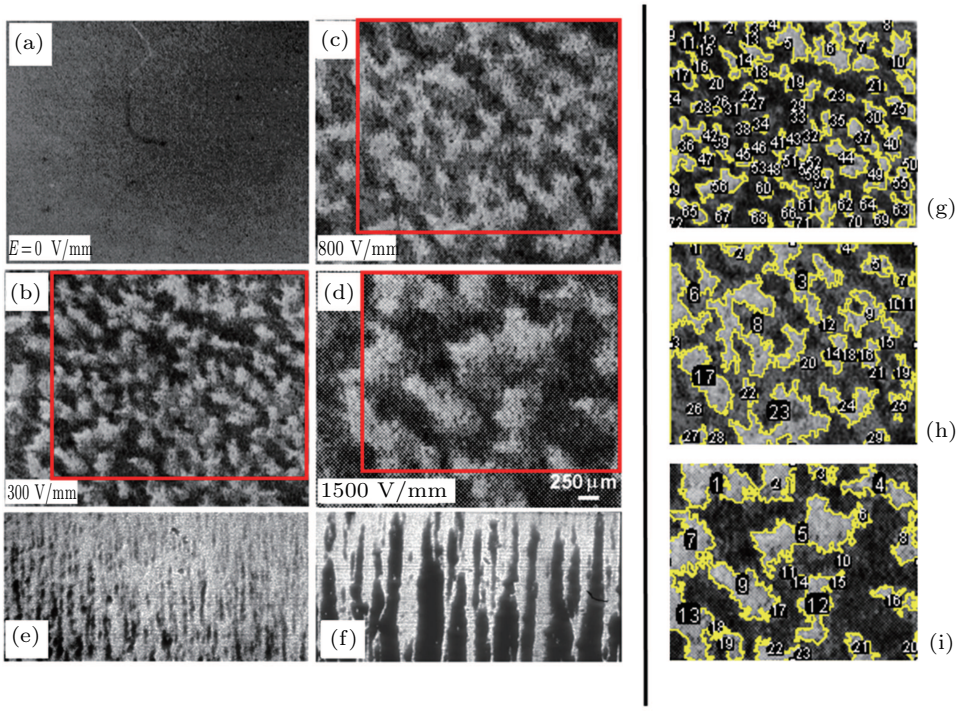

Fig. 1. (color online) Top and side views of ER fluid under different field strengths and top views after the software processing. The diameter of the glass microsphere is 0.5 μm and the volume fraction of the ER fluid is 0.15. Panels (a), (b), (c), and (d) have the same scale bar. ((a)–(d)) Morphologies of top views of ER fluid under 0, 300, 800, and 1500 V/mm, respectively; ((e)–(f)) Morphologies of side views of the ER structures respectively corresponding to panels (b) and (d); ((g)–(i)) parts of the red border in panels (b)–(d), which are processed by the software (ImageJ, National Institutes of Health), respectively. The yellow line indicates the column edge in the top view.

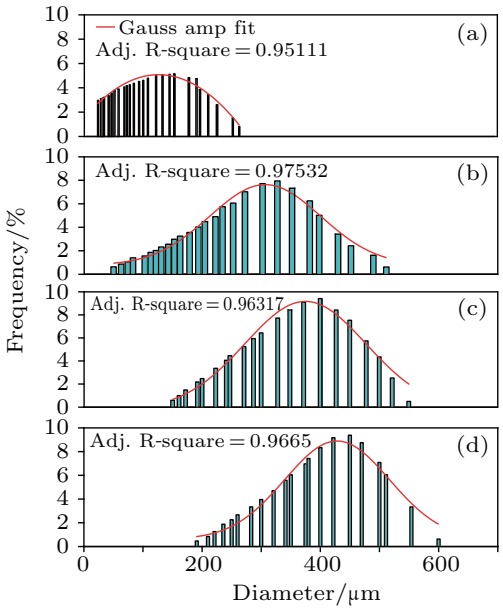

Fig. 2. (color online) Distribution profiles of column diameters observed under different electric field strengths. Diameter of the microsphere is 0.5 μm and the volume fraction of the ER fluid is 0.15. Red lines are the GaussAmp fit of the diameter distributions. ((a)–(d)) Electrical field strengths for the four graphs are 300, 500, 800, and 1200 V/mm, respectively. (a) When the electrical field strength is 300 V/mm, the diameters are in a range from 18 μm to 261 μm. The number of columns, whose diameters are distributed between 148 μm and 150 μm, is greatest. (b) When the electrical field strength is 500 V/mm. The diameters are in a range from 142 μm to 515 μm. The number of columns, whose diameters are distributed between 320 μm and 330 μm, is greatest. (c) When the electrical field strength is 800 V/mm, the diameters are in a range from 145 μm to 555 μm. The column number, whose diameters are distributed between 396 μm and 403 μm, is greatest. (d) When the electrical field strength is 1200 V/mm, the diameters are in a range from 188 μm to 603 μm. The number of columns, whose diameters are distributed between 446 μm and 453 μm, is greatest.

Figure 2 shows the size distributions measured at different field strengths, $\boldsymbol{E}$. We can observe that at $\boldsymbol{E} = 300$ V/mm, the columns are very thin fibrous structures [see Figs. 1(b) and 1(e)]. The coarsening process of the columns, driven by the dipole force among the particles, can be seen in Figs. 2(b) and 2(d) as $\boldsymbol{E}$ is increased. With the electrical field strength increasing, the distribution range of diameter increases. However, comparing Fig. 2(c) with Fig. 2(d) which are both under a high $\boldsymbol{E}$, their difference is not obvious. It reveals that the diameter is not linear with respect to $\boldsymbol{E}$. We also measure the distribution profiles when $\boldsymbol{E}$ surpasses 1500 V/mm but find no difference from the profile measured at 1500 V/mm. In our investigation, we also find that when $\boldsymbol{E}$ is greater than 500 V/mm, all the distribution profiles of the column diameters are fitted to a normal probability function.

Following Ref. [17], the mean diameter of column S_{d} as a function of electric field strength, $\boldsymbol{E}$, can be defined as

$$S_{\mathrm{d}}(E) = \frac{\sum n_s(\boldsymbol{E}) s^2}{\sum n_s(\boldsymbol{E}) s}, \tag{1}$$

where $n_s(\boldsymbol{E}) = N_s(\boldsymbol{E})/N_0$ with $N_s(\boldsymbol{E})$ being the number of columns with s chains (here the chain diameter is 0.5 μm) and N_0 the total number of chains. The sums are taken over all the columns. It is expected that

$$S_{\mathrm{d}}(E) \sim \boldsymbol{E}^Z \tag{2}$$

as $\boldsymbol{E}$ increases with z, a critical exponent.

Figure 3 shows the experimental results for the mean column diameter according to Eq. (1) for a field range of $\boldsymbol{E} = 500$ V/m–1500 V/mm. It can be clearly seen that the lower the field applied, the thinner the column diameter is. The dependence of the mean column diameter $S_{\rm d}(\boldsymbol{E})$, on field, $\boldsymbol{E}$, appears to approach to Eq. (2) with an exponent $z = 0.31 \pm 0.02$. The shear stress of the ER fluid, resulting from the field-induced interaction force among the particles, varies with mean column diameter. This is plotted in the inset of Fig. 3, where it is expected that the shear stress, τ, increases as mean column diameter, $S_{\rm d}(\boldsymbol{E})$, becomes larger and satisfies the power-law, $\tau \sim S_{\rm d}(\boldsymbol{E})^m$. By fitting the curve, it is found that the exponent, m, approximately equals 1.8 in the low field range. However, this is not the case when the field strength applied to the ER fluid surpasses a certain value. After that value, the shear stress increases even though the column diameter is saturated. The dipole-dipole force is probably strongly related to the distance between two microspheres.[4] Therefore, we believe that, in this case, the continuously increasing interaction among the particles must be caused by the slight adjustment in the gap between two nearly touching microspheres.

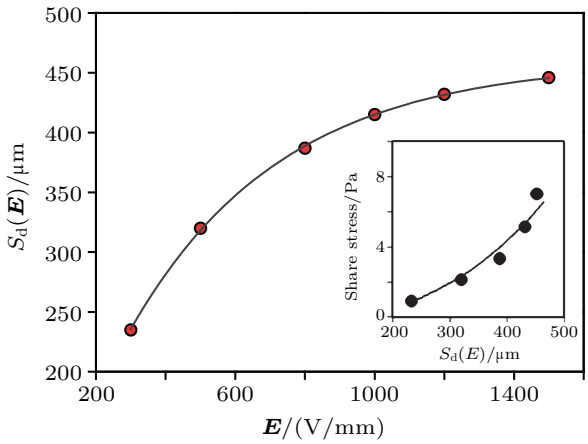

Fig. 3. (color online) Dependence of mean column diameter on electrical field strength. Inset shows the relationship between the shear stress and the column size. The mean column diameter has the exponential relation with $\boldsymbol{E}$ and the power is between 0 and 1. The shear stress of ER fluid is obviously proportional to mean diameter under a certain $\boldsymbol{E}$ to a particular power which is greater than 1, approximately.

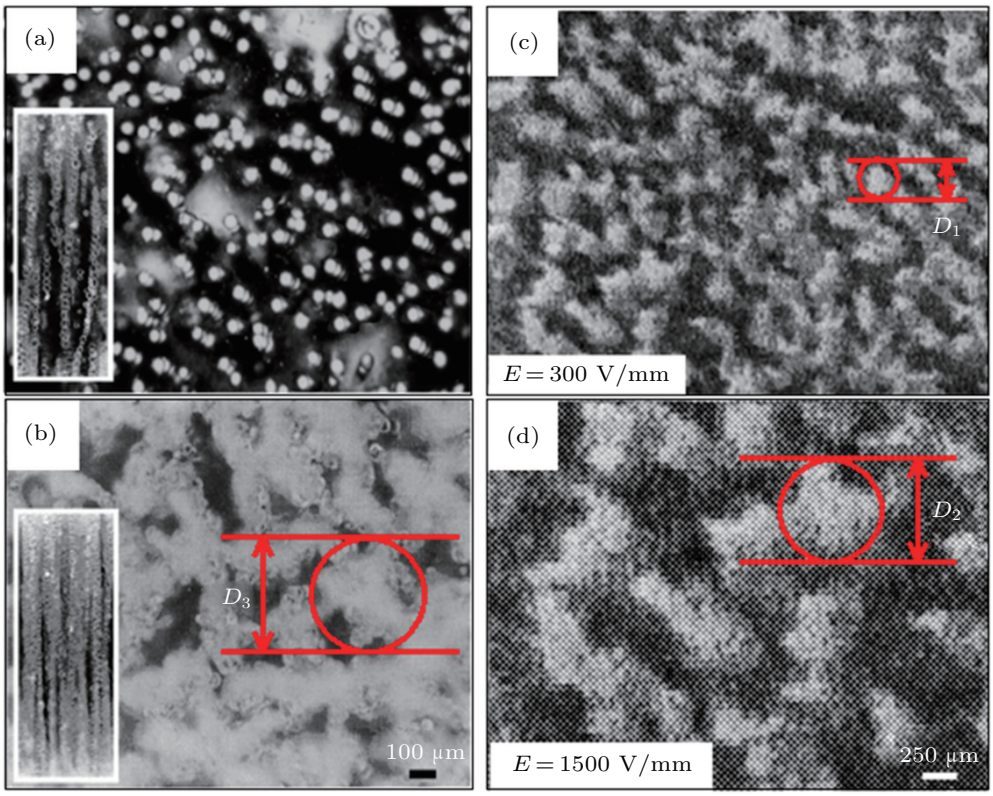

Fig. 4. (color online) Top views of ER suspension at different electrical field strengths with 0.5-μm or 47-μm glass microspheres. The volume fraction is 0.15. ((a) and (b)) Top views of ER suspension with 0.5-μm glass microspheres under 300 V/mm and 1500 V/mm, respectively. Insets are side views corresponding to each case. Two figures have the same scale bar; (a) one microsphere connects to another microsphere, forming a chain. As a result, the column diameter is the same as the diameter of microsphere; (b) $D_3 = 492.5$ μm. There is more than one microsphere per section; ((c) and (d)) Top views of ER suspension with 0.5-μm glass microspheres under 300 V/mm and 1500 V/mm, respectively. Two figures have the same scale bar, $D_1 = 261$ μm, $D_2 = 484$ μm.

The same aggregating phenomenon can be observed by replacing 0.5 μm microspheres with 47 μm ones as demonstrated in Fig. 4. From the top and side view images, the chains are spanned between two electrodes under low field strength, while the stable columns appear when the field strength was high enough. An interesting observation from our experiments is that with the same ER fluid cell, the saturated column diameter is almost the same no matter what the sizes of microspheres are. In the present case, the critical diameter of the column with either 0.5-μm or 47-μm microspheres is

about 500 μm. In Figs. 4(b) and 4(d), under an electrical field strength of 1500 V/mm, the D_2 is close to D_3. However, under the same low electrical field strength, the diameters in two ER fluids which are composed of 0.5-μm and 47-μm microspheres respectively are different (see Figs. 4(a) and 4(c)). In addition, we change the spacing between the two electrodes from 1.5 mm to 4 mm but do not find any obvious differences in the column diameter.

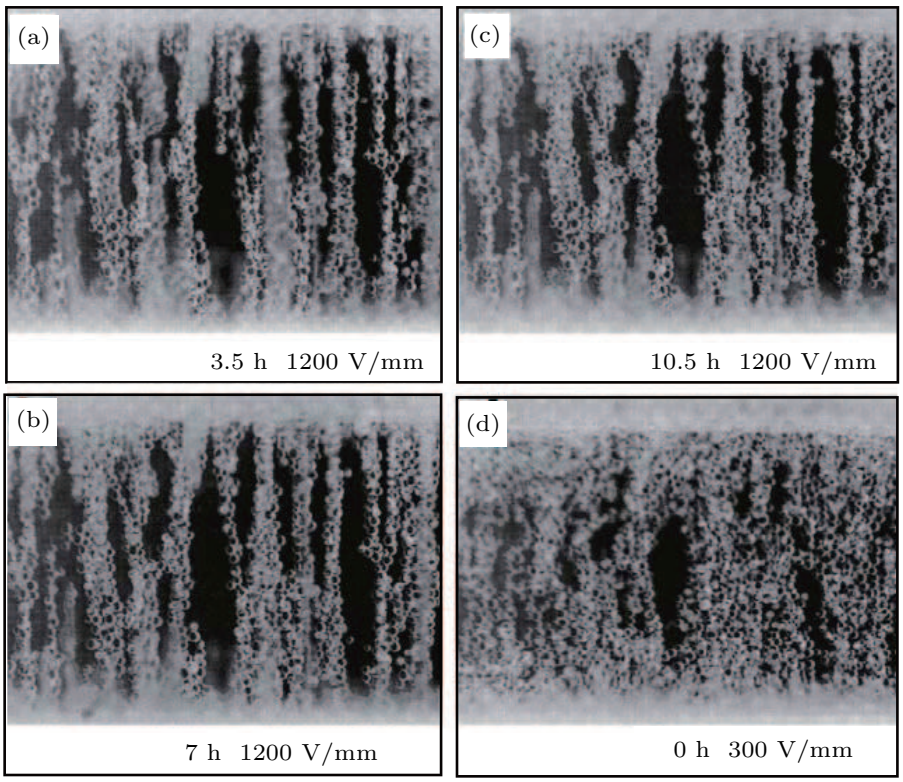

Fig. 5. Morphologies of ER fluid at different times, and field strengths. ((a)–(c)) Side view of the ER fluid at different times under 1200 V/mm. The column structures are stable even after 10.5 hours. (d) When the electrical field strength decreases to 300 V/mm, the columns are no longer suspended in the liquid and they sedimentate to the bottom of the cell.

Figure 5 shows the morphologies of columns at different times. At fixed field strengths of more than 1 kV/mm, the columns spanned between the two electrodes are very stable and no change occurs even after 10.5 hours (Figs. 5(a)–5(c)). However, when the field strength decreases to a critical value (300 V/mm), the columns are no longer suspended in the liquid and they sedimentate to the bottom of the cell, which can be seen in Fig. 5(d).

In conclusion, we have demonstrated experimentally that column ripening is strongly related to the force induced by the dipole-dipole interaction among dielectric particles in ER fluid. It is found that under a high field strength, there exists a critical column diameter that is not affected by the particle size. In addition, under a low field strength, the shear stress of the ER fluid increases and the mean column diameter becomes larger. This is not the case when the field strength surpasses a threshold value.

4. Conclusions and perspectives

The size distribution of columns in ER fluid strongly depends on the electrical field strength applied to the ER fluid, but the diameter of the column is not sensitive to the particle size. We observe that the interaction among particles increases exponentially as a function of column diameter when the ER fluid is under a low field strength. This is not the case when the electrical field strength is high. In addition, we find that the column diameter will be saturated when the field strength surpasses a critical value. The interaction among the particles, nevertheless, cannot be saturated even though the column size remains unchanged. For the future study, we will try other kinds of ER suspension dispersants and dispersed phases to investigate their microstructure transitions driven by the electric field. We will further analyze the effects of other external conditions on structural transition, such as temperature, volume fraction, etc.

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