Chain/column evolution and corresponding electrorheological effect

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(Received 21 August 1998; accepted for publication 2 October 1998)

We present an investigation about chain/column evolution and the corresponding electrorheological (ER) effect performed with glass/oil ER fluid. Our results demonstrate that once the field applied to the ER fluids surpasses a certain time period, the particles begin aggregating to form chains. These chains then coarsen and eventually form columns in the direction of the external field. We found that different column structures can be obtained depending on how the electric field is applied to the ER fluid. Only a loose column structure can be achieved if a square pulse field is applied to the ER fluid, yet a compact column is formed when the field strength is increased slowly. We have measured the ER effect with a sensitive yield stress testing device as the structure varies. The results indicate that there exist three increasing tendencies of interaction among particles corresponding to three processes of sequential transition between states; they are (1) random spatial configuration to chain, (2) chain to metastable column, and (3) metastable column to stable column. © *1999 American Institute of Physics.* [S0021-8979(99)07201-1]

I. INTRODUCTION

Electrorheological (ER) fluid is a kind of colloid which consists of dielectric particles immersed in an insulating liquid. It has a unique property that its viscosity increases dramatically upon application of an external electric field and returns to its original value when the electric field is removed. The change of viscosity of ER fluids is thought to be caused by the dipole–dipole interaction induced by the external field on the particles.[1–7] It is thought that the particles will aggregate to form chains and then to columns.[8] This phenomenon has recently been observed in magnetorheological (MR) fluids under a pulsed magnetic field.[9] For the ER fluid case, primitive studies about the chain formation[10] and structure evolution[11,12] have been carried out before, however, a direct observation of chain/column transition and the corresponding interaction among particles has not been explored.

In our previous paper,[13] we have shown that under a fixed strength of electric field, the water-free, watered, and metal coated microspheres in silicone oil will aggregate to form chains only when the acting time t_w (pulse-on time on the ER fluids) of the electric field on the ER fluid surpasses a certain threshold value. The chain formation time varies for different materials used. In this article, we present further experimental evidence of the particle aggregation processes. We found that the aggregating particles first form chains. Then the chains begin to coarsen and eventually form columns in the direction of the external field. We also found that only a loose column structure can be obtained if the square pulse field is applied to the sample, a compact column can be built if the field strength is increased slowly. We have measured the interaction among particles as the structure varies. The results indicate that there exist three steps of interaction strength corresponding to the transitions between three structures; they are (a) random spatial configuration to chain, (b) chain to metastable column, and (c) metastable column to stable column.

II. EXPERIMENTAL AND DISCUSSIONS

A. Evolution of chain/column transition–fast aggregation

The experimental cell is assembled by mounting two parallel electrodes on a glass slide with dimension of the cell to be 2.5 mm wide$\times$10 mm long$\times$5 mm high in which a drop of well-mixed ER fluid is dispersed. An optical microscope with video recorder is used to monitor the whole process of chain and column formation as the pulse-on electric field is applied to the sample. The period T of the pulse is fixed at 8.46 s (for the experimental setup see Ref. 13). The ER fluid used in the present experiment consists of water-free glass microspheres mixed with silicone oil. Figure 1 illustrates the aggregation process of glass microspheres under an external electric field pulse with $V_{p\text{-}p}$ of 1.2 kV/mm and variable power on time of t_w. To measure the minimum chain and column formation time of our system, we increased t_w gradually. The chain formation process shown in our previous paper,[13] at $t_w=0$ all the particles are dispersed randomly in the oil. Once t_w surpassed a certain value the particles formed chains suspended across the two electrodes. A series of images of the whole process of chain/column transition under two different field strengths is shown in Figs. 1(a)–1(d) and 1(e)–1(h), respectively. From Figs. 1(a) to 1(d), we notice that as t_w was increased, the chains suspended across two electrodes begin to coarsen and contact together to form thin columns. If t_w was further increased, the columns continue to coarsen and finally form thicker and

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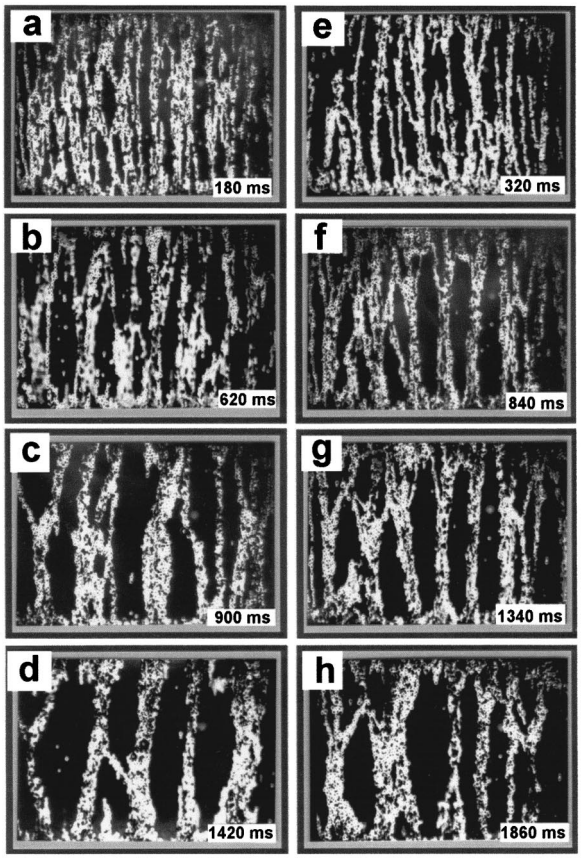

FIG. 1. Evolution of chain/column transition when the pulsed electric field is suddenly applied to the sample, where (a)–(d) and (e)–(h) are carried out under field strength of $V_{p-p}=1.2$ and 0.8 kV/mm, respectively. The volume fraction is 0.092.

more stable columns with finite diameters. The chain/column transition under a lower field strength was also tested and it was found that the weaker the field strength applied, the slower the column formed. This fact can be realized from Figs. 1(e)–1(h), where the minimum time for the chain and stable column formations are measured to be longer than 200 and 1400 ms, respectively. The average chain length and column width depend on the pulse on-time and this is illustrated in Fig. 2, where $\langle N\rangle$ is defined as the average number of particles per chain and $\langle N'\rangle$ is the number of chains per column in diameter. Regarding each data point, 16 chains and columns were counted for the particle and the chain numbers, respectively, and the average was taken. Only the chains and columns at the surface were enumerated, even though we found that the topography of the chains and columns did not vary with depth. It should be noted that the volume fraction of this ER fluid is at the medium value of 0.09, otherwise particle counting is difficult. Figure 2 indicates that $\langle N\rangle$ increases monotonically with t_w. A complete chain formed at 150 ms, after which the chains began to coarsen and $\langle N'\rangle$ increased. After 1.2 s, all columns were stable and $\langle N'\rangle$ was saturated. The column width in our system was less than 500 μm and the diameter-to-length ratio of a column was about 0.2.

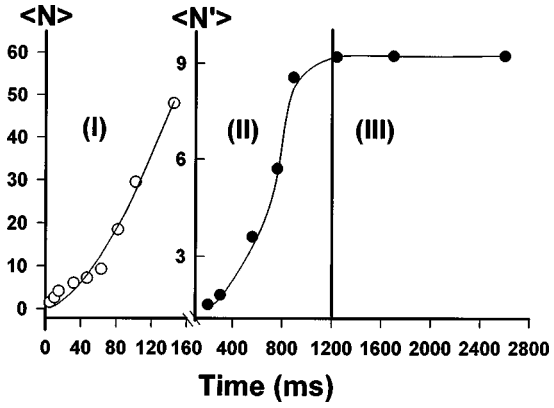

FIG. 2. Dependence of chain length and column width on the time scales, here $\langle N\rangle$ and $\langle N'\rangle$ are defined as average number of microspheres in a chain and average number of chains in one column. The regions I, II, and III represent different stages of structural evolution; they are; (I) From random configuration to chain (II), from chain to metastable column (III) from metastable column to stable column.

B. Evolution of chain/column transition–slow aggregation

We noticed that all the columns observed in Fig. 1 are loose structures and it seems that no ordered structure could be built. However, according to the theoretical prediction, the lowest energy state of ER fluid is a stable state at which all particles should aggregate to form ordered structure, i.e., in most cases the body centered tetragonal (BCT) structure.[1–3,14] We realized that if the rise time of the electric field is short or the field is applied suddenly, the particle's movement is so fast that it cannot reach its lowest energy state, which means that the state presented here might not be in equilibrium. In order to verify our hypothesis, an additional experiment was carried out by applying the electric field to the sample linearly and slowly (the rate of increase of field strength was fixed at 30 V/mm per second). The whole process of structure evolution is shown in Fig. 3, where we

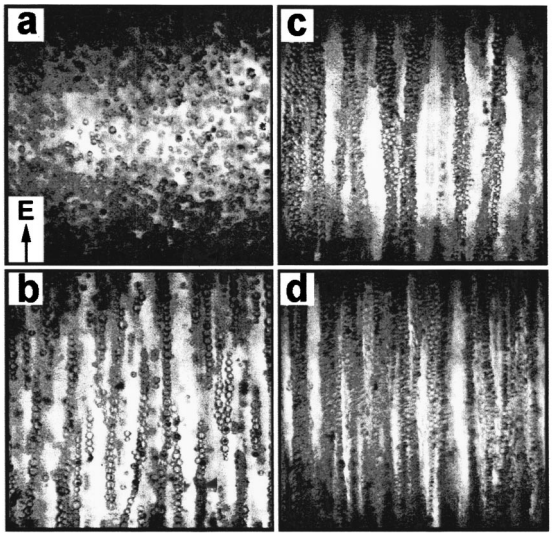

FIG. 3. Evolution of chain/column transition when a dc electric field is increased very slowly. The field strengths for (a), (b), (c), and (d) are 0, 500, 900, and 1200 V/mm.

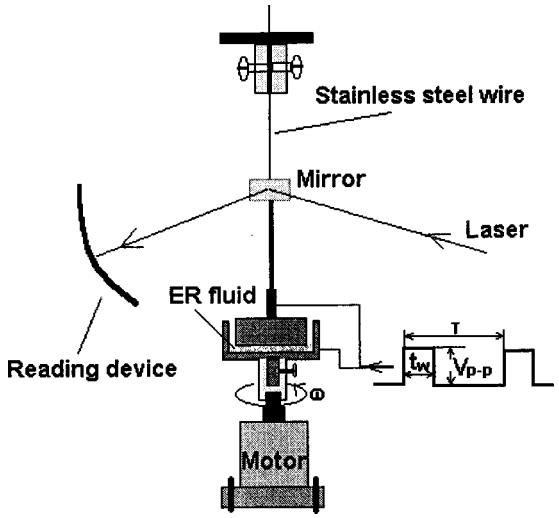

FIG. 4. Setup for ER effect measurement. The motor speed is 0.001 rpm.

can see that while the field strength was increased to 500 V/mm, all suspended microspheres aligned themselves to form complete chains across the two electrodes, Fig. 3(b). Thin columns can be seen when the field strength reached 900 V/mm, Fig. 3(c). The columns continued to coarsen and eventually formed stable columns even when the electric field was further increased, see Fig. 3(d). Here we note that the columns are compact structures. However, it should be pointed that in the present experiment, it is difficult to determine the column structure. In fact, for most applications of ER fluids, the external electric field is applied to the system within a very short time. In this case, it is very difficult to form any ordered structure as mentioned above. However, the ideal model for the theoretical calculation may be based only on the latter case, that is, the aggregation of particles proceeds slowly enough so that the compact structure of column formed has sufficient time to equilibrate.

We would like to emphasize here that, in our systems, the column diameters formed by glass microspheres are limited even if we further increased the electric field. As an example of the electric-field-induced system, according to Ref. 12, for a one-dimensional structure, a dipole chain or column will experience strong Landau–Peierls fluctuation and the aggregation of new particles to the chains or columns will take place preferentially at the tip. This will serve as an upper limit on the column width. The expected columns will have a radius $r_{\perp} \sim a(L/a)^{2/3}$, where L and a are the length of the column and the radius of a microsphere, respectively. In the present case, $L=2.5$ mm and $a=25$ μm. The column width in our system can be calculated to be about 1.07 mm which is twice the experimental value. We believe that there is competition between the repulsive and attractive forces between two chains or columns. The larger the column becomes, the slighter the fluctuation of the column. This might be the reason for the presence of a critical column diameter in our system.

C. Time response of ER effect

In order to measure particle interaction as structures changed, a sensitive ER effect testing device was designed and the experimental setup is shown in Fig. 4. As can be seen from this figure, the ER effect was measured by using a parallel plate torsion device. The motor rotating speed is fixed at 0.001 rpm during the entire testing process. With a pulsed electric field applied across the ER fluid sandwiched between the two parallel plates, the lower plate was mounted to a motor whose rotating speed could be controlled. The upper plate was held by a stainless wire on which a mirror was fixed. Without electric field, the zero position on the reading device was adjusted when the lower plate was rotated very slowly, dragging the top plate. As an electric field was applied, the viscosity of ER fluids increased and resulted in the deviation of the mirror from its original position. The information on the screen could then be read out until slipping occurred between the two plates. Our testing result is given as Fig. 5. We found that the time response of ER effect changes with particle size. The trend shows that the smaller the particle size, the shorter the ER effect response time. Under the same experimental conditions, the initial response time for ER fluid with 1.5-μm-diameter particles is shorter than 5 ms, whereas the ER fluid consisting of 47-μm-diameter particles is near 100 ms. One would imagine that the larger the particle size used, the longer the response time for ER effect. Using a 1.5 μm sample, at $t_w=500$ ms, the dependence of the ER effect on the magnitude of the pulse field is determined as shown in the inset of Fig. 5. The test is performed by increasing the field strength $V_{p\text{-}p}$ from 500 to 2500 V/mm while measuring the variations of T_x, which is defined as the time needed for the laser spot to move 20 divisions in distance. It can be seen that T_x decreases as field strength is increased, which means that the higher the field strength the stronger the ER effect.

A more interesting phenomenon, which can be seen in Fig. 5, is the existence of three increasing steps of ER effect. In order to compare the ER effect with corresponding structure changes, we use the example of 47-μm-diameter microspheres. We found that the first step lasted from 60 to 160

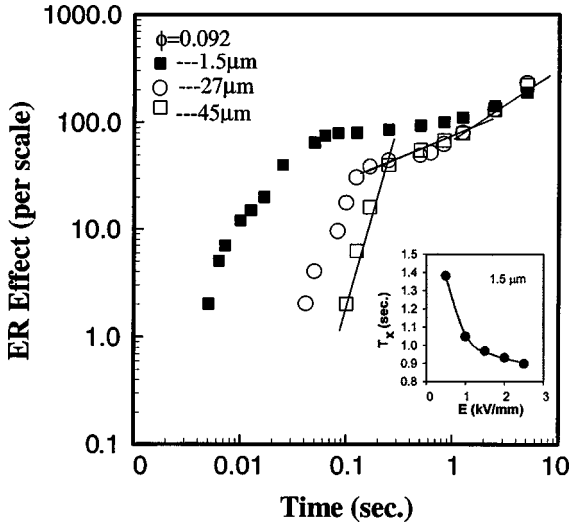

FIG. 5. Dependence of ER effect on the acting time to the sample. Inset shows field dependence of ER effect at fixed $t_w=500$ ms. Here T_x is the time that the laser spot on the reading device moves 20 divisions (here each division is 2.5 mm in distance).

ms, the second from 160 ms to 1.2 s, and the third corresponded to time after 1.2 s, which is consistent with the three structural changes, (1) random spatial configuration to chain; (2) chain to metastable column; and (3) metastable column to stable column. Unfortunately, the chain/column transition for 1.5 μm particles could not be exhibited here due to difficulty in observation of the small particles under a normal optical microscope.

III. SUMMARY

In situ observation of the chain/column transition performed with water-free glass microspheres suspended in silicone oil have been seen under an external time-regulated electric field. It was found that the chain/column transition occurred once the acting time t_w on the sample surpassed a certain value. Only a loose column structure can be achieved if the electric field is applied suddenly, while the compact column is obtained when the applied field strength is increased very slowly. The diameter of columns observed in the glass microspheres/oil colloid is finite and less than 500 μm. In addition, we have also measured the particle interaction as the structure varied. The results show that there exist three increasing tendencies of interaction among particles which correspond to three processes of the structure changes.

[1] L. C. Davis, Appl. Phys. Lett. **60**, 319 (1992); Appl. Phys. Lett. **81**, 1985 (1997); Phys. Rev. A **46**, 719 (1992).
[2] T. C. Halsey and W. Toor, Phys. Rev. Lett. **65**, 2820 (1990).
[3] H. Ma, W. Wen, W. Y. Tam, and P. Sheng, Phys. Rev. Lett. **77**, 2499 (1996).
[4] H. J. H. Clercx and G. Bossis, Phys. Rev. E **48**, 2721 (1993).
[5] M. Whittle, W. A. Bullough, D. J. Peel, and R. Firoozian, Phys. Rev. E **49**, 5249 (1994).
[6] H. Conrad, in *Particular Two-Phase Flow*, edited by M. C. Roco (Butterworths, Boston, MA, 1992), p. 355.
[7] A. Khusid and A. Acrivos, Phys. Rev. E **52**, 1669 (1995).
[8] R. Tao, Phys. Rev. E **47**, 423 (1993).
[9] J. H. E. Promislow and A. P. Gast, Phys. Rev. E **56**, 642 (1997).
[10] J. C. Hill and T. H. Van Steenkiste, J. Appl. Phys. **70**, 1207 (1991).
[11] D. J. Klingenberg, C. F. Zukoski, and J. C. Hill, J. Appl. Phys. **73**, 4644 (1993).
[12] J. E. Martin, J. Odinek, and T. C. Halsey, Phys. Rev. Lett. **69**, 1524 (1992); Phys. Rev. E **57**, 756 (1998).
[13] W. Wen, D. W. Zheng, and K. N. Tu, Phys. Rev. E **57**, 4516 (1998).
[14] T. J. Chen, R. N. Zitter, and R. Tao, Phys. Rev. Lett. **16**, 255 (1992).