Dynamics of disklike clusters formed in a magnetorheological fluid under a rotational magnetic field

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We investigate the cluster formations and dynamics in a magnetorheological fluid under a rotational magnetic field focusing on the case of a relatively high volume fraction. We find that isotropic disklike clusters, which rotate more slowly than the field rotation, are formed at low Mason numbers (the ratio of viscous to magnetic forces) and, what is more, we show short rod clusters, which rotate stably thanks to the low Mason numbers and circulate along the surface of the disklike clusters. The circulation velocity of the surface particles is much higher than the rotational surface velocity of the rigid disklike clusters.

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Magnetorheological (MR) fluids are colloidal solutions, in which paramagnetic particles of 0.1–10 μm in diameter are dispersed. When an MR fluid is subjected to a dc magnetic field, a magnetic dipole moment is induced in each particle and the magnetic particles form chain clusters in the direction of the external field via the dipole-dipole interactions [1–3]. If the external field is rotated, the chain clusters rotate following the field rotation. The dynamics of chain clusters in rotational magnetic fields has been investigated in detail experimentally, theoretically, and numerically [4–11]. Now it is well known that rotating chain clusters are deformed by hydrodynamic drag and then become S-shaped. As the frequency increases, the clusters are finally broken into shorter chain clusters. The chain clusters become shorter with an increase in the frequency and those short clusters coagulate to form isotropic disklike clusters on the plane of the rotating magnetic fields [6–8,12–15]. The important parameter, which governs the dynamics and structures of clusters formed in MR fluids under rotational magnetic fields, is the Mason number, that is, the ratio of viscous drag to magnetic forces acting on a particle,

$$\mathrm{Ma} \equiv \frac{12^2 \mu_0 \eta \omega}{M^2}, \tag{1}$$

where μ_0, η, ω, and M are, respectively, the magnetic permeability, the viscosity of a solvent, the angular velocity, and the magnetization of a magnetic particle. It was shown that in the case of low volume fractions ranging from 10^{-4} to 0.02, the critical Mason number, at which chain clusters break up into isolated particles and isotropic clusters are formed, is approximately 1, irrespective of the volume fractions [8]. In the case of higher volume fractions, on the other hand, chain clusters coagulate in the lateral direction and thick chain clusters are formed [1], and therefore the structures and dynamics of clusters may be different from those in the case of low volume fractions. The rotational motion of rigid disklike clusters, which is caused by the phase lag between external rotational electric fields of extremely high frequencies and the induced dipole moments (so-called rotary ER effect), was investigated and the dependence of the rotational motions of the disklike clusters on the intensity and frequency of a rotational electric field was clarified [16]. However, the cluster structures and dynamics formed in an MR fluid of relatively high volume fractions under a rotational magnetic field have not yet been studied in detail. It is very important to clarify the cluster formation and dynamics in external fields not only from a scientific point of view but also from a practical point of view since they can be utilized in the fields of nano/microtechnology, biochemistry, biotechnology, bioengineering, etc. [17–27]. In this Brief Report, we investigate the formations of isotropic disklike clusters and the dynamics of paramagnetic particles in an MR fluid of a relatively high volume fraction under a rotational magnetic field.

The outline of our test cell and experimental system is shown in Fig. 1. The MR fluid used in this study is composed of paramagnetic particles (M-PVA C22, Chemagen Co.), which are dispersed in water. The M-PVA C22 paramagnetic particles are made of polyvinylalcohol, in which magnetite grains are suspended. The average diameter of the particles, which we measured by digital image analysis, is 1.82 ± 0.59 μm. The MR fluid was confined between two glass substrates. The depth of the MR fluid layer was set at 4 μm using nonmagnetic spacer particles [see Fig. 1(a)]. The volume fraction of the particles in the present MR fluid was 0.081, which is much higher than that in the previous work [8], where the critical Mason number for the breakdown of chain clusters and the formation of isotropic clusters was clarified for a volume fraction of 10^{-4} to 0.02. We confirmed from the images of particles taken during the experiment that the clusters formed in the test cell were two-dimensional. The area fraction of the paramagnetic particles in the test cell, which was measured from the projection to the plane, which is parallel to the particles' layer, was 0.22. A rotational magnetic field was produced by coils, function generators, and amplifiers [see Fig. 1(b)]. The sequence of the experiment is shown in Fig. 2. At first, the MR fluid was placed in a dc magnetic field for 30 min and then the magnetic field was rotated. As is shown in Fig. 2, the frequency of the

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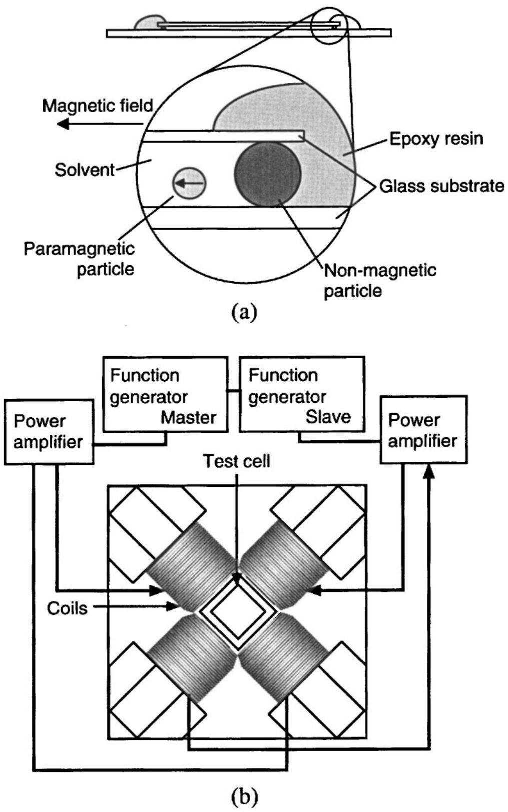

FIG. 1. Schematic representations of test cell (a) and experimental system (b).

rotational magnetic field was increased stepwise from 0.1 to 10 Hz. Note that the intensity of the magnetic field was constant at 12.7 kA/m throughout the whole experiment. In this case, the average induced magnetic dipole moment in a particle is 3.0×10^{-20} Wb m. We define the nondimensional parameter λ, which is the ratio of the dipole-dipole interparticle potential energy to thermal energy,

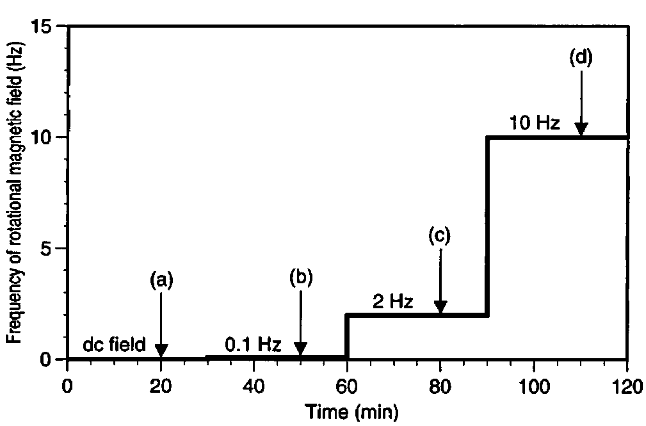

FIG. 2. Experimental procedure. The frequency of a rotational magnetic field was increased stepwise. The intensity of the field was constant at 12.7 kA/m.

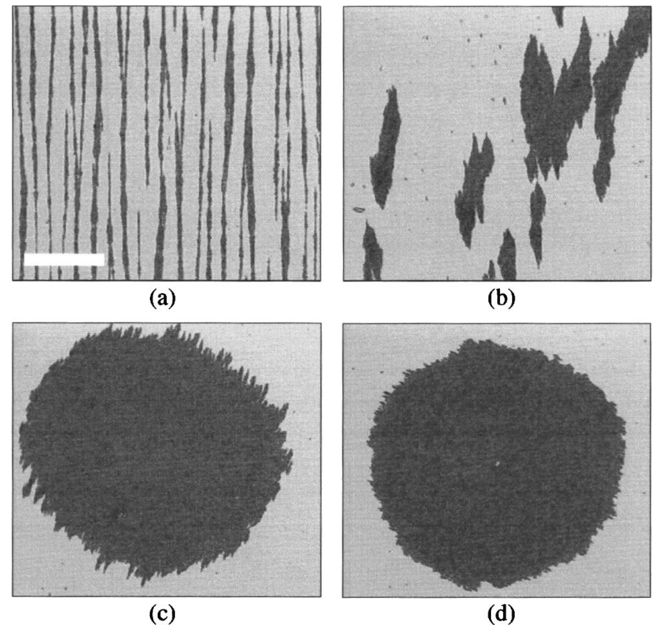

FIG. 3. Cluster structures of paramagnetic particles in dc and rotational magnetic fields. (a) dc magnetic field, (b) 0.1 Hz (Ma $=1.3\times10^{-3}$), (c) 2 Hz (Ma$=2.5\times10^{-2}$), (d) 10 Hz (Ma$=0.13$). The snapshots (a)–(d) are, respectively, obtained at the points (a)–(d) indicated in Fig. 2. The inset scale bar is 100 μm.

$$\lambda \equiv \frac{|\mathbf{m}|^2}{4\pi\mu_0 d^3 kT}, \qquad (2)$$

where $\mathbf{m}$, d, k, and T are, respectively, the dipole moment vector of a particle, the diameter of a particle, the Boltzmann constant, and the temperature. The average value of λ in the experiment was 2279. The cluster dynamics was observed by a charged-couple-device (CCD) high-speed video camera, which was connected to a computer, and was recorded on both videotape and the hard disk of the computer.

The snapshots of clusters formed in the dc and rotational magnetic fields are shown in Fig. 3, where the pictures were taken at the points (a)–(d) indicated in Fig. 2. In the dc magnetic field, chain clusters were formed along the field [Fig. 3(a)]. When the frequency of the rotational magnetic field was as low as 0.1 Hz (Ma$=1.3\times10^{-3}$), chain clusters rotated following the magnetic field rotation. Some of the rotating chain clusters collided with each other in the lateral direction since the volume fraction of the present MR fluid is relatively high, and, as a result, thicker rod clusters were formed [Fig. 3(b)]. When the frequency was increased to 2 Hz (Ma$=2.5\times10^{-2}$), the rod clusters dissociated into shorter rod clusters, which finally aggregated to form isotropic disklike clusters [Fig. 3(c)]. Here, let us point out that the critical Mason number, at which the transition in the cluster structures from anisotropic to isotropic ones occurs, is much smaller than 1. Rotating clusters were broken into shorter ones immediately after the frequency was changed from 0.1 Hz to 2 Hz. Those short clusters coagulated to form larger clusters since the volume fraction is quite high as we mentioned, and eventually disklike clusters were formed. Now, in order to understand the formations of disklike clusters, let us compare the time scales of the rotations of the induced dipole moments and the Brownian translational motions of particles. The Brownian relaxation time of a colloidal particle is

$$\tau_B \equiv \frac{3\pi\eta R^3}{kT}, \tag{3}$$

where R is the radius of a particle. Supposing that the rotation of the dipole moments follows the field rotation, the ratio of the Brownian relaxation time to the period of the dipoles' rotation becomes 0.17, 3.4, and 17.1 in the cases of 0.1, 2, and 10 Hz, respectively. It is quite apparent that isotropic disklike clusters are likely to be formed rather than rod clusters once the angular velocity of the rotational field or the dipole moments becomes higher than the translational motions of particles. In such high-frequency regions, the effective interparticle potential energy between the particles can be considered as the dipole-dipole potential energy averaged over one cycle of the rotational field, which is isotropic and attractive [16,28]. Therefore, once isotropic disklike clusters are formed in such high-frequency rotational fields, they remain stable.

Next, let us focus on the dynamics of the particles, which formed the disklike clusters. Figures 3(c) and 3(d) show disklike clusters of similar size formed in the cases of 2 and 10 Hz [29]. The disklike clusters rotated slowly. The angular velocity of the disklike clusters was much lower than the frequency of the rotating field. For instance, the angular velocities of the clusters shown in Figs. 3(c) and 3(d) were 0.055 and 0.103 rad/s, respectively. The most significant features concerning the disklike clusters are the surface roughness and the movement of the surface particles. The motions of the particles near the surface are shown in Fig. 4 [29], which corresponds to Figs. 3(c) and 3(d). Since the values of the Mason number are, respectively, 2.5×10^{-2} and 0.13, which are smaller than 1, in the cases of 2 and 10 Hz, the surface particles can form rod clusters stably and the clusters can rotate following the external field. The surface in a frequency of 10 Hz was smoother than that in 2 Hz since the surface rod clusters were shorter in the former case than those in the latter case. It was clearly observed that short rod clusters rotated and circulated along the surface, while the inner particles hardly moved. We measured the circulation velocities of the surface particles [see Figs. 4(a) and 4(b)]. The average circulation velocities at 2 and 10 Hz were, respectively, 45.8 and 57.7 μm/s, which were much faster than the surface velocity of the rigid disklike clusters. The average circulation speed of the surface rod clusters V_c is determined by the frequency f and length L of the clusters: $V_c=2fL$. Expressing the dependence of the cluster length on the frequency as in the case of low volume fractions, $L\sim f^{\Delta}$, V_c is expressed as follows: $V_c\sim f^{1+\Delta}$. According to a theory [9], experiments [5,6,8], and numerical simulations [5,8] for low volume fraction systems, $\Delta=-0.5$ and, therefore, $V_c\sim f^{0.5}$, while in the present relatively high volume fraction case, the value of the power $1+\Delta$ may be lower than 0.5 since the dependence of L on f is different from that in low volume fraction cases due to the formation of the thick rod clusters. In fact, if we estimate the average length of the surface rod clusters from the trajectories of the surface particles, it is 14.7 and 4.6 μm in the case of 2 and 10 Hz, respectively, which suggests $\Delta\sim-0.7$ and therefore $V_c\sim f^{0.3}$. We will be evaluating precisely the power $1+\Delta$.

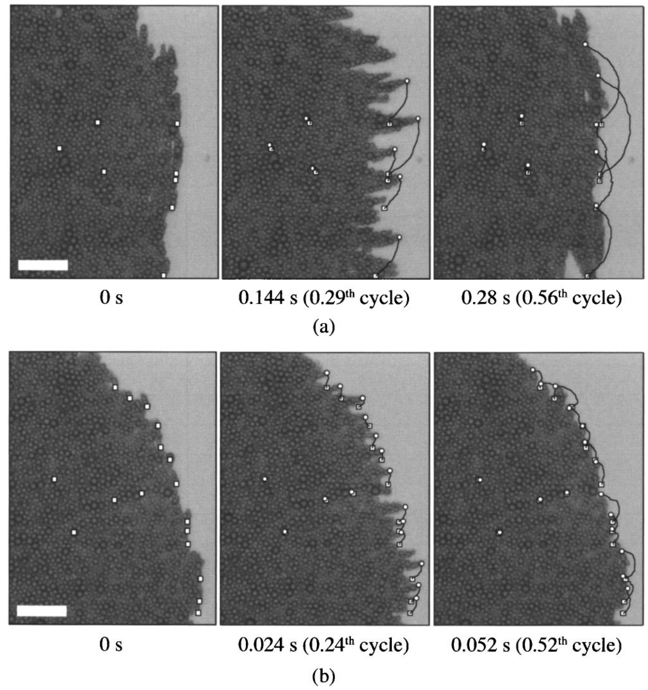

FIG. 4. Motions of particles along the surface of disklike clusters. (a) 2 Hz, (b) 10 Hz. The magnetic field was rotated anticlockwise. The solid curves shown in the figures are the trajectories of the particles we focused on. Symbols (□) and (○) represent the starting point and the snapshot position of each particle at the indicated time. Whereas the inner particles hardly moved at such a short time interval, the surface particles formed short rod clusters and these clusters rotated following the field rotation and circulated along the surface. The inset scale bar is 20 μm.

In summary, we studied the cluster formations and dynamics in an MR fluid of a relatively high volume fraction under a rotational magnetic field experimentally using optical microscopy. We clarified the dependence of the cluster structures on the frequency of the external magnetic field. We found that disklike clusters are formed as the frequency of the external field increases, and we observed the rod clusters rotating and circulating along the surface of the disklike clusters. These specific features in the formation and dynamics of the rod clusters along the surface of the disklike clusters appear only when the Mason number is smaller than 1, in which case rod clusters can stably rotate along the surface, and the volume fraction is high, in which case disklike clusters are formed even when $\mathrm{Ma}<1$. In order to understand the dynamics of both disklike and surface rod clusters quantitatively, we need to estimate the shear stress and torque acting on the clusters, which we will be investigating in detail. This rod clusters' movement along the surface also suggests that magnetic particles can be manipulated along a magnetic substrate by applying a rotational magnetic field and, what is more, the direction of the manipulation can be controlled by changing the rotational direction of the magnetic field.

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[29] See EPAPS Document No. E-PLEEE8-71-056503 for movies corresponding to Figs. 3(c) and 3(d) and Figs. 4(a) and 4(b). A direct link to this document may be found in the online article's HTML reference section. The document may also be reached via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html) or from ftp.aip.org in the directory/epaps/. See the EPAPS homepage for more information.