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Flow Behavior and Microstructure Developed between Parallel Plate Electrodes for Electro-Rheological Nano-Suspensions under No External Electric Fields

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The flow behavior of nano-suspensions with particle volume fractions of $\phi = 2.4$ and 4.4 vol% was investigated under no electric fields using a rotational rheometer with fixtures of parallel plates (or electrodes), the gap distances of which were narrower than several hundred micrometers. For the sample of $\phi = 4.4$ vol%, the apparent yield stress was observed with gap distances of 50 μm and 300 μm . On the other hand, the flow behavior was strongly dependent on the gap distance for the sample of 2.4 vol%. These results were discussed in terms of the size of the microstructure and the gap distance between the parallel plates of the rheometer.

Key Words: Narrow gap / Nano-particles / Yield stress / Microstructure / Electro-rheology

1. INTRODUCTION

The electro-rheological (ER) effect is the reversible rheological responses of fluids only by application and removal of an external electric field. A suspension composed of micro-particles and insulating oil is known to show the ER effect.¹⁾ The flow of the ER micro-suspension under no electric fields is generally assumed to be the Newtonian flow. Under an electric field, the flow is well assumed to be the Bingham flow. Because a characteristic response time of the ER micro-suspension is on the order of milliseconds^{2,3)}, there are substantial expectations for practical applications⁴⁻⁶⁾, such as dampers, clutches, valves, robotics, force display devices, and so on.

For practical applications, however, there are still demands for the ER fluid to be improved. Although a much higher yield stress has been a major demand, stability in the ER effect would also be a demand. Furthermore, applicability for a narrow gap between the electrodes is required for micro- or nano-devices. Therefore, particular attention has been directed toward a suspension based on nano-particles.⁷⁾

Recently, a suspension based on dry titanium dioxide (TiO_2) nano-particles with the nominal average diameter of the primary particles of 15 nm has been reported.⁷⁻⁹⁾ The suspended particles were remarkably stable against

sedimentation and electrical breakdown. With the nano-suspension, a gap of 50 μm between the parallel plates of the rheometer (or electrodes) was easily filled. In a previous paper⁹⁾, the steady shear flow behavior of nano-suspensions with different volume fractions of TiO_2 nano-particles (ϕ) was investigated under no external electric fields. Unlike the ER micro-suspension, the apparent yield stresses^{10,11)}, which were measured even under no electric fields with a gap distance of 50 μm between the parallel plates of the rheometer, were found for the samples of $\phi = 2.4$ and 8.8 vol%. On the other hand, no plateau region was found for the sample of $\phi = 1.2$ vol%. These results, some of which are shown for comparison in Figs. 1 and 2 of this paper, were discussed in terms of the microstructure composed of secondary particles. It is suggested that the attractive and repulsive forces among nano-particles of TiO_2 were not so strong but the attractive one predominated¹⁰⁾, because the dried nano-particles with no specific stabilizations were suspended in a dried silicone oil. Consequently, secondary particles or further aggregated microstructure was developed. The microstructure developed macroscopically and spread throughout the parallel plates will be responsible for the apparent yield stress. It was proposed that randomly distributed secondary particles or a macroscopically developed microstructure of secondary particles in the nano-suspension should also be taken into account for the initial state of the ER effect, other than randomly distributed primary particles in the ER micro-

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suspension generally assumed. For applications to micro-flow devices, the flow behavior within gap distances narrower than several hundred micrometers should also be evaluated in relation to the microstructure of the nano-suspension.

In the present study, the flow behavior within gap distances narrower than several hundred micrometers was investigated under no electric fields using a parallel-plate rheometer for nano-suspensions of $\phi = 2.4$ and 4.4 vol%. Optical observations of the microstructure after shear deformations were also performed with two levels of magnification, which were lower than the previously reported magnification⁹⁾ because the width of the imaging field of view was limited to around 50 μm . Therefore, the microstructure, which was observed using a much wider imaging field of view, can be discussed based on the gap between the parallel plates of the rheometer.

Fig. 1. The shear stress plotted against the shear rate for the nano-suspensions with particle volume fractions of $\phi = 2.4$ and 4.4 vol%. Previously reported data⁹⁾ were also plotted in the figure.

Fig. 2. The shear stress plotted against the shear rate for the nano-suspension of $\phi = 2.4$ vol% measured with $h = 50$ μm and 100 μm . Previously reported data⁹⁾ were also plotted in the figure.

2. EXPERIMENTAL

Rutile TiO_2 nano-particles with the nominal average diameter of the primary particles of 15 nm were suspended in a silicone oil with the viscosity of 5×10^{-2} Pa·s. The volume fractions of the particles were $\phi = 2.4$ and 4.4 vol%. The details of the preparation of the samples were similar to that previously reported.⁷⁻⁹⁾

The rheological measurements were performed under no electric fields at room temperature using a strain-controlled rotational rheometer (Rheology Co., Ltd., MR-300V2E) and fixtures of the parallel plates (electrodes). In the present study, stability of the torque sensor has been improved against pressure drop synchronized with the air-compressor in our rheometer. The diameter of the plates (D) and the gap distance between the plates (h) were chosen, respectively, from 18 mm to 30 mm, and from 30 μm to 500 μm . The data measured with $D = 30$ mm and different gap distances of h will be reported unless otherwise stated. Before each measurement, the sample was pre-sheared at a constant shear rate of 200 s^{-1} for 30 s, and kept in the quiescent state for 300 s. Particle behavior was observed in the quiescent state using an optical microscope (Nikon, Eclipse E600W) equipped with a CCD camera. A small amount of the sheared nano-suspension was carefully mounted on a glass slide and optically observed. Typically, the upper plate of the fixture was removed for sampling, and the glass slide was carefully contacted with the nano-suspension on the upper and lower plates. In some cases, the nano-suspension on the plate was directly observed. During the sampling, the nano-suspension was elongated and compressed to same extent. It can be thought that the macroscopically developed microstructure was broken into smaller fractions, if the microstructure was fragile, and that the broken fractions were also re-combined. However, it was confirmed from observations that the microstructure spread around 50 μm or 100 μm was preserved after careful sampling.

3. RESULTS AND DISCUSSION

3.1 Steady Shear Flow Behavior of the Nano-Suspensions

Figure 1 shows the shear stress plotted against the shear rate for the samples of $\phi = 2.4$ and 4.4 vol% measured with $h = 50$ μm . The data previously reported⁹⁾ are also plotted for comparison for the samples of $\phi = 8.8$ vol% (which were measured with $D = 18$ mm and $h = 50$ μm), 2.4 vol% ($D = 30$ mm, $h = 50$ μm), and 1.2 vol% ($D = 40$ mm, $h = 300$ μm). For the sample of $\phi = 4.4$ vol%, a plateau stress around 2 Pa is

found at the lower shear rates. In the present paper, the plateau stress is regarded as the apparent yield stress.⁹⁻¹¹⁾ The apparent yield stress of the sample of $\phi = 4.4$ vol% is slightly higher than that of the sample of $\phi = 2.4$ vol% (around 1 Pa), but much lower than that of the sample of $\phi = 8.8$ vol% (around 10 Pa). The apparent yield stress σ_{y0} measured with $h = 50$ μm showed a dependence slightly stronger than ϕ . The exponent was not determined here partly because scattering of the data is found in Fig. 1 for the sample of $\phi = 2.4$ vol% (This Work), and also because a plateau region was not always found in the present study for the sample of $\phi = 2.4$ vol% especially for the wider gap distances of h .

Figure 2 shows the shear stress plotted against the shear rate for the nano-suspension of $\phi = 2.4$ vol% measured with $h = 50$ μm and 100 μm . Previously reported data⁹⁾ were also plotted. For the sample of $\phi = 2.4$ vol% measured with $h = 100$ μm , no plateau region is found in Fig. 2. It should also be pointed out that the torque responses were rather weak for the sample of $\phi = 2.4$ vol% measured with $h = 300$ μm , so that the flow curve could not be obtained using our rheometer. In a few cases, no plateau region was also found for samples of $\phi = 2.4$ vol%, which were separately prepared and measured with $h = 50$ μm , while a plateau region was found with $h = 30$ μm . These changes in the flow behavior due to the gap distance of h were especially observed for the sample of $\phi = 2.4$ vol% in the present study. Figure 3 shows the shear stress plotted against the shear rate for the sample of $\phi = 4.4$ vol% measured with $h = 50$ and 300 μm . Similar to the previously reported results⁹⁾ for the sample of $\phi = 8.8$ vol%, a plateau region is clearly seen not only for the flow curve measured with $h = 50$ μm , but also for that measured with $h = 300$ μm . The apparent yield stress measured with $h = 300$ μm is slightly lower than that measured with $h = 50$ μm . It is suggested that the flow behavior is closely related to the size of the microstructure developed between the parallel plates.

3.2 Optical Observations of Microstructure

Figure 4 shows optical micrographs taken in the quiescent state for the sheared samples of $\phi = 2.4$ vol%. The micrographs of (a) and (b) were taken after the shear deformations discussed in Figs. 1 and 2 with $h = 300$ μm , and those of (c) and (d) with $h = 100$ μm . The micrographs of (e) and (f) were taken after shear deformations with $h = 50$ μm , in the flow curve of which a plateau stress around 1 Pa was observed. The white portions in the micrographs represent secondary particles (or particle rich phase), while the black portions represent the silicone oil (or oil rich phase). The white portions may be slightly enhanced in the micrographs

because of multiple scattering of light, overlapping of particle rich phase, and some inclusions of the silicone oil to the phase. A macroscopically developed microstructure of secondary particles was previously found⁹⁾ for a sample of $\phi = 2.4$ vol%, the microstructure of which looked like a network formed by

Fig. 3. The shear stress plotted against the shear rate for the nano-suspension of $\phi = 4.4$ vol% measured with $h = 50$ μm and 300 μm .

Fig. 4. Optical micrographs for the nano-suspension of $\phi = 2.4$ vol% after the shear deformations discussed in Figs. 1 and 2 with $h = 300$ μm , (a) and (b), and with $h = 100$ μm , (c) and (d). The micrographs of (e) and (f) were taken after shear deformations with $h = 50$ μm , in the flow curve of which a plateau stress around 1 Pa was observed.

the secondary particles. The network-like microstructure was significantly spread beyond the imaging field of view, but the width of the imaging field of view was around 50 μm . In the present study, the microstructure was also observed using a much wider imaging field of view. In Fig. 4, the length scale of 50 μm (or 300 μm) is shown for comparison. Secondary particles and part of a further aggregated microstructure, which is related to a network-like microstructure⁹⁾, are seen in (a), while the entire microstructure can be seen in (b). In (a), an aggregated microstructure of secondary particles is well spread around 50 μm . On the other hand, it is suggested that the microstructure in (b) could not be densely developed enough throughout the gap distance of $h = 300 \mu\text{m}$, so that the microstructure was separately distributed. Although a weakly connected network-like microstructure may be seen because of the multiple scattering, overlapping, and so on, boundaries of many aggregates of the particle rich phase can also be found in (b). Such a fragile microstructure with possible boundaries would be closely related to the weak responses of torque for $h = 300 \mu\text{m}$. Characteristics similar to (a) and (b) can be found, respectively, in (c) and (d). In (c) and (d), a microstructure was developed slightly larger than the gap distance of $h = 100 \mu\text{m}$. As shown in Fig. 2, no plateau region was still found with the gap distance, but stress responses were detected at the higher shear rates, suggesting some defects or boundaries in the microstructure.

Furthermore, characteristics similar to Figs. 4 (a) and (b) can basically be found, respectively, in (e) and (f) which were taken after shear deformations with $h = 50 \mu\text{m}$, but also a network-like microstructure is more pronounced in (e). The network-like microstructure is well spread beyond the length scale of 50 μm and boundaries or defects would be reduced within the scale, with the gap distance of which the plateau region was observed in Fig.1. In (f), boundaries of aggregates of the particle rich phase can be found. However, the microstructure in (f) also appears to be densely developed, suggesting that the microstructure sheared with $h = 50 \mu\text{m}$ was different from the microstructure of (b) or (d). For the samples prepared separately and sheared with $h = 50 \mu\text{m}$, the microstructure similar to (b) or (d) was also observed. Further studies are needed to make clear the effect of gap distance of h suggested above. It is also considered that the size of the microstructure was slightly distributed, especially for the sample of $\phi = 2.4 \text{ vol}\%$, so that scattering of the rheological data was sometimes observed.

For the sample of $\phi = 4.4 \text{ vol}\%$, the characteristic microstructure, which would be responsible for the plateau region shown in Figs. 1 and 3, was not seen with a sufficient

Fig. 5. Typical optical micrographs for a sample of $\phi = 4.4 \text{ vol}\%$ after shear deformations with $h = 300 \mu\text{m}$.

contrast. In a micrograph of the sample, the white portions were remarkably enhanced. A micrograph similar to that of $\phi = 4.4 \text{ vol}\%$ was also reported⁹⁾ for the sample of $\phi = 8.8 \text{ vol}\%$. Figure 5 shows typical optical micrographs taken in the quiescent state for the sample of $\phi = 4.4 \text{ vol}\%$ after shear deformations with $h = 300 \mu\text{m}$. (The micrographs taken after shear deformations with $h = 50 \mu\text{m}$ (not shown here) and with $h = 300 \mu\text{m}$ were similar.) The white portions are widely spread beyond the length scale of 300 μm in Fig.5 (b). Therefore, a possible microstructure was densely developed beyond the gap distance of $h = 300 \mu\text{m}$, so that an apparent yield stress was measured with the gap distance. Further study will be undertaken to understand the ER behavior of the samples with different initial microstructures under no electric fields.

4. CONCLUSIONS

In the present study, the flow behavior of nano-suspensions of $\phi = 2.4$ and $4.4 \text{ vol}\%$ under no electric fields was discussed in terms of the size of the microstructure and the gap distance between the parallel plates of the rheometer. For the sample of $4.4 \text{ vol}\%$, the apparent yield stress was observed with gap distances of 50 μm and 300 μm . On the other hand, the flow behavior was strongly dependent on the gap distance especially for the sample of $2.4 \text{ vol}\%$. These results were closely related to the size of the microstructure developed between the parallel plates in comparison to the gap distance.

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