Floc Structure and Flow Properties of Pulp Fiber Suspensions

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The rheological properties of pulp and PET fiber suspensions were measured with a parallel-plates type rheometer. A high speed CCD camera was used to observe the changes in the floc structures that were produced during the rheological measurement. The flow curves of the pulp fiber suspensions showed Newtonian flow in the low shear rate range. With increasing shear rate, the shear stress increased and then became unstable, namely, jumps in the flow curves were observed, which are due to the formation of fiber flocs. At a higher shear rate, the flocs disappeared and the systems showed Newtonian flow again. The flow curves of the polyethylene terephthalate (PET) fiber suspensions showed non-Newtonian flow even in the low shear rate range. We found that the uniform distribution of fibers became uneven and then the flocs began to form at the critical shear rate; and over the critical shear rate, the fibers began to uniformly disperse again, i.e., the flocs disappeared. The two-dimensional fast Fourier transform (FFT) technique and Guinier approximation were used to obtain the radius of gyration, \( R_g \), of the flocs. It is considered that the \( R_g \) of the flocs is useful to characterize the mechanism of fiber flocculation.

Key Words : Fibers / Flocculation / Suspensions / Image analysis / Radius of gyration

1. INTRODUCTION

The floculation of fibers in water is a widely encountered phenomenon. For example, pulp fibers are stored, transported, and processed in water throughout the operations of pulp and paper manufacture. The floculation of pulp fiber occurs in flowing suspensions. The fibers within the suspensions are mostly distributed unevenly, giving local mass concentrations called flocs. The formation of flocs in the pulp suspensions leads to heterogeneity and poor formation in paper. Therefore, in the common papermaking process pulp fiber suspensions with concentrations as low as 1% are required to obtain a uniform flowing system. It is important to understand the floc formation mechanism and floc behavior in the flow field for controlling the flow properties of the fiber suspension.

Recently, the effects of fiber length, fiber concentration, fiber curl, fiber aspect ratio on the pulp flocculation and floc size have been studied.\(^1\)\(^-\)\(^3\)\) However, there have been few studies about the floc, especially under dynamic conditions at high flow rate. This is because the flocculation process is difficult to experimentally observe, as fibers are small, opaque, and moving rapidly in most applications.\(^4\)

The fiber suspension shows complicated rheological properties because of the orientation and the interaction between the dispersing fibers and also the formation of fiber networks in the flow field. The network structure of the fibers leads to a solid-like property, i.e., the yield stress and plateau modulus of the fiber suspension.\(^5\)\) Moreover, the flow property became more complicated due to the formation of flocs. The state of flocculation is strongly related to the flow property of fiber suspensions.

In this study, the rheological properties of the pulp fiber and polyethylene terephthalate (PET) fiber suspensions of various concentrations were measured. The flocculation of the fiber suspensions was observed with a high speed CCD camera. The two-dimensional fast Fourier transform (FFT) technique was used to obtain the power spectral patterns from the obtained images, and then according to the Guinier approximation\(^6\), the radii of gyration, \( R_g \), of the flocs were calculated and the state of the flocculation was quantitatively evaluated.

2. EXPERIMENTAL

2.1 Materials

A commercially available hardwood bleached kraft pulp fiber was used in this study. The average length and diameter of the fibers were 1.1 mm and 15 \( \mu \)m, respectively. As a comparison, we also used polyethylene terephthalate (PET) fiber, TK04N, kindly supplied by Tenjin Co., Ltd., Japan. The length and the fineness of TK04N were 3 mm and 0.1 denier;
i.e., 3.2 \mu m in diameter. These fibers were dispersed in distilled water to obtain suspensions of various concentrations.

2.2 Instrumentation and measurements

A modified Weissenberg R-18 Rheogoniometer (Sangamo Controls, Ltd., UK) equipped with a pair of Petri dishes as parallel plates was used. The diameter of the dishes and the gap between them were 152 mm and 3.9 mm, respectively. The rheological properties of pulp fiber and PET fiber suspensions were measured at 25°C. The shear rates ranged from \(1.2 \times 10^1\) to \(1.2 \times 10^2\) s\(^{-1}\) in these measurements.

A high speed CCD camera (fastcam-net, Photron, Ltd., Japan) was used to capture the images of the flowing fiber suspensions. Each image was in gradations of 256 gray levels and was taken every 1/250 s. Their picture elements are 480 \times 512 pixels. The camera was fixed near the edge of the dishes so that the images can be captured at the same position where the motions of the fibers were clearly observed. The size of original images were 24 \times 26 mm.

According to the Guinier approximation, the radius of gyration, \(R_g\), can be calculated from the scattering intensity as shown in Eq. (1), which always can be fitted to any particles\(^6\), irrespective of the particle shape.

\[
I(k) = I(0) \exp \left(-k^2R_g^2/3\right)
\]  

(1)

Here, \(I(k)\) is the scattering intensity and \(k\) is the spatial frequency. The formula is fitted if \(k\) is very small. All calculations should be done under the condition of \(R_g^2k^2 < 1\), though the suitable scopes for the application of Eq. (1) are slightly different depending on the specific shapes of the particles. For example, it was found that the condition is \(R_g^2k^2 < 1.3^2\) for the normal spherical particle. Thus, using the Guinier plots in which the abscissa is \(k^2\) and the ordinate is \(\ln(I(k))\), one can calculate \(R_g\) from the slope.

In this study, a two-dimensional fast Fourier transform (2DFFT) technique was used to obtain \(I(k)\) and \(k\). Namely, we took the two-dimensional power spectral patterns (PSP) from the original high-speed images using 2DFFT. It was executed using a window diameter of 440 pixels and in gradations of 256 gray levels. Each pixel corresponds to 0.05 \times 0.05 mm\(^2\) in real space. The intensity of the PSP was calculated by the circular average from the center.

3. RESULTS AND DISCUSSION

3.1 Flow properties

Figure 1 shows the logarithmic plots of the shear stress, \(\sigma\), versus the shear rate, \(\gamma\), at various pulp fiber suspension concentrations. In the low shear rate range up to \(1.8\) s\(^{-1}\), the flow curves show Newtonian flow. As the shear rate increases, the flow curves display some discrete jumps in shear stress, which suggests an unstable state. This is due to the floc formation of the dispersing fibers in the flow field as described below. The value of the shear stress of a relatively highly concentrated suspension jumped at the lower shear rate than that of the dilute one. When the shear rate continued to increase, ranging from \(5.8 \times 10^1\) to \(1.2 \times 10^2\) s\(^{-1}\), the jumps disappeared, and the flow curves showed Newtonian behavior again. The flow curve of the 0.4% suspension showed a plateau in the higher shear rate range.

Figure 2 shows the flow curves of PET fiber suspensions having various concentrations. These flow curves are different

![Figure 1](image1.png)

*Fig.1 Logarithmic plots of shear stress, \(\sigma\), versus shear rate, \(\gamma\), for various concentrations, \(c\), of pulp fiber suspensions.*

![Figure 2](image2.png)

*Fig.2 Logarithmic plots of shear stress, \(\sigma\), versus shear rate, \(\gamma\), for various concentrations, \(c\), of PET fiber suspensions.*
from those of the pulp fiber suspensions. They show non-Newtonian flow even at the relatively low concentration of 0.03%. With increasing shear rate or concentration, the shear stress obviously increased. However, in the range from 1.8 to $2.9 \times 10^1$ s$^{-1}$, the shear stress is unstable, which is similar to the case of the pulp suspensions. The ranges in which the jumps of the shear stress can be seen were narrower than those of the pulp fiber suspensions for the same concentration. This jump in shear stress is attributed to the floc formation of the PET fibers as described below. In the lower shear rate region, the flow curves of the PET fiber suspensions are straight lines. Their slopes decreased with increasing concentrations. Comparing the flow curves of the pulp and the PET fiber suspensions at the same concentration, we found that the shear stress of the flow curves of the PET fiber suspensions is higher than those of the pulp fiber suspensions for most of the shear rate range. Furthermore, the PET fiber suspensions show non-Newtonian flow. These facts should be due to the fact that the PET fiber is longer and thinner than the pulp fiber.

3.2 Observations of fiber flow and flocculation

Figure 3 and 4 show the images of the pulp fiber and the PET fiber suspensions under different shear rate conditions, respectively. Here, the fiber concentration is 0.05% so that it can be easily observed. The alphabetical marks correspond to the same ones in the flow curves as shown in Fig.1 and Fig.2. From Fig.3a, we found that the pulp fibers disperse well and are likely to uniformly flow. Figure 3b shows the image at the shear rate when the relative movements of the interfiber are predominant and flocculation of the fibers begin. The floc size at this stage was small. With increasing shear rate, the interfiber movement then becomes more active, which leads the fibers to form more flocs as shown in Fig.3c. When there is some interference between the fibers, being assembled is more
stable for fibers than being separated. In the flowing systems, the relatively large flocs act as independent bodies in the flow field. Figure 3d shows the image at the shear rate point where the flocs dispersed again and the suspension turned more uniform. One of the reasons for these interesting phenomena is considered to be due to the centrifugal force and velocity gradation in the floc. The higher the shear rate, the larger becomes the force and the velocity gradation, so that it breaks the flocs. Mason postulated that flocculation is a dynamic equilibrium process with fibers continuously moving in and out of the flocs, both rates being equal at the steady state. In this study, similar phenomena were observed in the pulp fiber and the PET fiber suspensions, i.e., the system expressed Newtonian flow again at this higher shear rate range.

The images shown in Fig. 4 also describe the flocculation process of the PET fiber suspensions at the different shear rates. This phenomenon is similar to that of the pulp fiber suspensions. The flocs of the PET fiber suspensions are larger than that of the pulp fiber suspensions, since the former is longer and thinner than the latter. The surface tensions of the pulp and PET are 42 mN/m and 44.3 mN/m at 25°C, respectively. Therefore, the effects of the surface properties of these fibers on the flocculation were ignored.

3.3 Analysis of floc structure

In the shear rate range from 1.8 to 2.9 × 10¹ s⁻¹, the flocculation noticeably occurred and the flocculation is very complicated as described above. The shape and size of the flocs were varied with the flow conditions. Understanding this flocculate process and changes in the flocculation at various shear rates is very important in the processing fiber suspensions.

The shape of the floc is vague and irregular, so that it is difficult to quantitatively estimate its size. Therefore, an

Fig. 4 Images of 0.05% PET fiber suspension at various shear rates. Alphabetical marks correspond to those shown in Fig. 2. Arrow indicates the flow direction for each image. Bar: 5 mm.
Fig. 5  (a) PSP of the image of 0.05% pulp fiber suspension shown in Fig.3d. (b) Logarithmic plot of the intensity, $I(k)$, of PSP versus spatial frequency, $k$. It is obtained from the circular average of Fig.5a.

Fig. 6  Images of a PET fiber floc at different values of threshold in 256 gray levels. (a) Original; (b), (c), and (d) are binarized whose threshold values are 15, 105, and 185, respectively. Bar: 5 mm.
evaluation method for the floc size is required. We tried to evaluate the floc size using 2DFFT of the images.9-11) Here PSPs correspond to the light scattering patterns.9) Figure 5a is the PSP image in Fig.3d. Figure 5b shows a logarithmic plot of the intensity, $I(k)$, versus the spatial frequency, $k$, calculated from the circular average of Fig.5a. These figures include much information concerning the floc; for example, the shape, the orientation, the interference between fibers and flocs, and so on. Thus, we need to separate the information of the floc size from the figures. Image processing is required to simplify the images as we expected, for example, the binarization of the images.

Figure 6 shows the original and binarized floc images. The binarization was used to change the apparent size of a floc, which can be controlled by changing the threshold values. The threshold values gradations of 256 gray levels are shown in Fig.6. The larger the threshold value, the smaller the apparent floc size is formed. From these images, the PSPs were obtained by 2DFFT. The radius of gyration, $R_g$, of the flocs obtained from the Guinier approximation 6) was used for evaluating the floc size.

Figure 7 shows the Guinier plots calculated from the PSPs of Fig.6. This figure includes $R_g$ calculated from the slope of the straight lines of the plots using Eq. (1). It is indicated that with the increasing value of threshold, $R_g$ decreased. The $R_g$ values obtained were 2.00 mm, 1.88 mm, and 1.43 mm for the threshold values of 15, 105, and 185, respectively. The $R_g$ obtained from the original image was 1.84 mm, which corresponds to that obtained in the threshold value of 105. The value of $R_g$ of a floc looks smaller than its apparent size,
because the $R_g$ of a thin disk is equal to $1/\sqrt{2}$ of its radius in three dimensions according to the morphological relationship. In conclusion, the combined 2DFFT and Guinier plot techniques are useful for characterizing the floc size if we correctly select the threshold value. It can also be stated that $R_g$ from the Guinier plot is useful to check the validity of the binarization. Considering the suitability for all images, we selected the threshold of 128 as the standard value in the following experiment. The refractive indices of the pulp and PET are 1.54 and 1.58, respectively.8) We considered that this very slight difference in the refractive indices did not affect the binarizing.

Most of images included many flocs and their boundaries are ambiguous. Therefore, we processed Fig.3d, and its binarized image is shown in Fig.8. Because the Guinier approximation is only applicable for a single particle system6), the $R_g$ of the flocs should be calculated one by one using image processing. Thus, these flocs in Fig.8 were separated from each other, and then each floc formed a new image, which can avoid any interference between the flocs and fibers in the FFT process. These new images were individually transformed by FFT, then the $R_g$ of each floc was calculated from Eq. (1) as shown in Fig.9. We can obtain the quantitative average $R_g$ value of 1.80 mm, which is considered as the characteristic floc size of this image. We have captured the images in flowing conditions at various shear rates, so the characteristic floc sizes at each shear rate can be calculated in this way. The averaged $R_g$ values were then used to evaluate the state of the flocculation of the fiber suspensions under flowing conditions.

The effects of the shear rate on the average radii of gyration, $R_g$, of the flocs are shown in Fig.10. When the shear rate increased, the $R_g$ also increased. For the shear rate ranging from 3.7 to $1.5 \times 10^1$ s$^{-1}$, the systems are very unstable and flocculation is noticeable as shown in Fig.3c and Fig.4c, so that the flocs are too large to calculate their $R_g$ value using the present method. In the higher flow rate region, $R_g$ decreases, which indicates a decrease in the flocculation. The $R_g$ of the flocs of the PET fiber suspension is greater than that of the pulp fiber suspension at various shear rates as shown in Fig.10. It is suggested that the flocculation of the PET fiber suspension is more noticeable than that of the pulp fiber suspension and this is due to the larger axial ratio of the PET fibers.

4. CONCLUSIONS

In the lower shear rate range, the shear stress of the fiber suspensions increased with shear rates. With increasing shear rate, there are some discrete jumps in the flow curves due to the unstable conditions by the floc formation. When the shear rate increased, the jumps disappeared. This means that the fibers disperse again and the systems show a Newtonian flow. The observations of the images using a high speed CCD camera correspond to the unique rheological properties of the fiber suspensions because of their flocculation.

With the 2DFFT technique, the PSP can be obtained from the high-speed images. According to the Guinier approximation, $R_g$ can be easily obtained from the PSP. The threshold values in the binarization can be evaluated using the obtained $R_g$ of the flocs, which is a useful tool for characterizing fiber flocculation regimes.

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