Turbulence characteristics and mixing performances of viscoelastic fluid flow in a serpentine microchannel

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Abstract. Flow velocity measurement and visualization using particle image velocimetry and fluorescent dye were carried out for a viscoelastic fluid flow in a serpentine microchannel for the purpose to quantitatively evaluate the unsteady flow characteristics that is observed even under very low Reynolds number regime due to the combined effect of the viscoelastic fluid properties and the channel shape. Sucrose water solution (Newtonian fluid) and the polyacrylamide-sucrose water solution (viscoelastic fluid) were used as working fluids. The mixing performance markedly increased when the Reynolds number exceeded a certain value in the polyacrylamide solution case. The single-point, cross-sectional and two-dimensional velocity distributions showed that low frequency fluctuation was produced in the polyacrylamide solution case. Particularly large fluctuation in the channel spanwise direction was observed in the upstream area of the serpentine channel. On the other hand, the amplitude of the fluctuation decreased in the downstream region. The fluctuation in the upstream region is believed to be generated by the flow instability at the curved part of the channel, while the fluctuations in the downstream area were attributed to the local instability and the vortices provided from the upstream region.

1. Introduction

Viscoelastic fluids are now well known to produce a significant difference in the flow characteristics from Newtonian fluids in terms of reducing turbulence and producing drag reduction effects in high Reynolds number flow regime (James, 2009), or increasing the flow instability in low \(Re\) regime (Burghelea et al., 2004; Larson et al., 1990). Recently, Groisman, A. & Steinberg, V. (2001), and Burghelea et al. (2004) reported that by adding polymers of several hundreds of ppm to the fluid, the flow instability is enhanced and flow fluctuation is produced in a \(mm\)- or \(\mu m\)-scale serpentine channels due to the elasticity of the fluid.

Instability study and experimental measurements have been keenly carried out in the past few decades on the flow characteristics of viscoelastic fluids in Taylor-Couette and Taylor-Dean flows (Larson et al., 1990; Joo, Y. L. & Shaqfeh, E. S. G., 1994). However, the flow structure, especially the turbulence characteristics of the viscoelastic fluid flow in serpentine channels have not been studied and understood yet. The study of such flow structures can give us an insight for designing an effective channel shape to be used in micro fluidic devices or compact mixing and heat transfer equipments, and provide large contributions to the fields of chemistry, heat transfer and medicine.

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In this study, velocity measurements using micro particle image velocimetry (µPIV) and visualization using fluorescent dye are carried out for a viscoelastic fluid flow in a serpentine microchannel in order to investigate the turbulent flow characteristics and mixing performances quantitatively. Polyacrylamide solution is used as viscoelastic fluids and is compared with a sucrose solution which is a Newtonian fluid. The influence of the curvature and streamwise position in the serpentine channel on the fluctuation characteristics, and the two-dimensional distributions are particularly discussed in this paper.

2. Experimental procedure and conditions

2.1. Experimental apparatus
The visualization and PIV measurement, which will be described shortly, were carried out using a microchannel and invert-type microscope. The objective lens was a long working distance lens with ×40 magnification and numerical aperture $NA=0.75$ (Olympus Co.; UPLFLN40XPH). The images were recorded by a high speed digital video camera (Vision Research; Phantom V7.3). The CCD resolution was 800×650 pixels, and the optical resolution of the image in combination with the lens was 0.542 µm/pixel. The focusing depth of the image was 1.7 µm.

The schematic of the microchannel is shown in Fig. 1. The channel had two inlets which join at the Y-shape junction. At the downstream of the junction was positioned the serpentine channel where the measurement was carried out. The serpentine part of the channel consists of 14 periodical circular curve units. The inner and outer radials $R_i$ and $R_o$ of the curve part were 100 and 180 µm. The channel width and height $W$ and $H$ were 84 and 60 µm, respectively. The microchannel was made of PDMS (poly-dimethylsiloxane) and was prepared by using SU-8 (MicroChem Co.) as a casting mold that was fabricated through a lithography process. The PDMS channel removed from the SU-8 was attached to a cover glass. The working fluids were supplied to the inlet of the microchannel by syringe (Hamilton) and syringe pump (Nihon Kohden Co.; CFV-3200) through a Teflon tube.

Two types of working fluids were used in this study. The first one was a 64.4wt% sucrose water solution. The sucrose solution was prepared by adding sucrose (nacalai tesque Co.) to ultrapure water purified by Direct-Q UV (Millipore Co.), and was mixed by rotating the container (As One Co.: AV-1) at 250rpm for 12hours. This fluid was regarded as a Newtonian fluid and is referred to as $\text{\textcopyright sucrose solution}$ hereafter. The other one was a water solution of 64.4wt% sucrose, 1wt% NaCl, 100ppm Polycrylamide (Polysciences Inc.: Polycrylamide 18522-100, molecular weight=18,000,000). The polymer solution was mixed under the same condition of the sucrose solution. This fluid is regarded as a viscoelastic fluid and is referred to as $\text{\textcopyright PAAm solution}$ hereafter.

2.2. Visualization measurement
Visualization measurement was carried out by mixing fluorescent dye in the working fluid and measuring the fluorescent intensity distributions. 3wt% RhodamineB (Kishida chemical Co.; E27036) was mixed with the solution and was used as fluorescent dye. The fluorescent solution was supplied from one of the inlet, and a solution without fluorescent mixture was supplied from
Table 1. Measurement conditions.

| $Q$ [$\mu l/min$] | $U_m$ [m/s] | $Re$ | $Wi$ |
|------------------|-------------|------|------|
| 0.08~1.6         | $2.7 \times 10^{-4}$~$5.3 \times 10^{-3}$ | $1.8 \times 10^{-4}$~$3.6 \times 10^{-3}$ | 17.6~352 |

the other side. Mercury lamp (Olympus Co.: U-LH100HGAP) was used as the light source to produce the excitation light that was guided to the microchannel through a band-pass filter (Olympus Co.: 25BP530), dichroic mirror (Olympus Co. BA575IF), and the objective lens. The fluorescence images of the dye was recorded by the high speed camera through another band-pass filter (Olympus Co.: BA575IF) in order to filter out the excitation and background lights. The sampling rate and exposure time of the camera were set as 100Hz and 70$\mu$s, respectively.

2.3. PIV measurement

Fluorescent micro-particles (Molecular Probes Co.: F8819, nominal diameter= 1.0$\mu$m) were mixed in the fluid as tracer particles in the PIV measurement. The same optical system employed in the visualization measurement was used in the PIV measurement. In order to measure a pair of images with appropriate time difference $\Delta t$ that is required for the PIV analysis, the high speed camera was triggered by pulse signals generated by a function generator (NF Co.: WF1974). Further, the sample rate of these pair images $f_s$ was appropriately selected to capture the low frequency fluctuation of the flow field. $\Delta t$, $f_s$ and the sample number of pair images were set as, 250$\mu$s, 100Hz and 1250, respectively.

2.4. Experimental conditions

The experimental conditions for the visualization and PIV measurements are summarized in Table 1. The flow rate was changed in the visualization measurement, so that the Reynolds number $Re = U_m D_h / \nu$ varied in the range of $1.8 \times 10^{-4}$~$3.6 \times 10^{-3}$. $U_m$ and $D_h$ are the cross-sectional mean velocity and the hydraulic diameter. The Weissenberg number, $Wi = 4\lambda U_m / D_h$, in this case ranges from 17.6 to 352. In the PIV measurement, on the other hand, the condition of $Re = 2.2 \times 10^{-3}$ only was considered following the results obtained by the visualization.

3. Results and Discussion

3.1. Visualization results

To provide some information on the visualization pattern recorded in the measurement, instantaneous photographs of the fluorescence distributions measured at the streamwise locations of curve $N = 2, 7$ and 13 in the case of PAAm solution is shown in Fig. 1. In the sucrose case, the boundary between the fluid with and without fluorescent dye formed along the streamwise direction at the middle of the channel was clearly observed at the $N=2$ curve. This boundary continuously remained in the downstream area to the $N=13$ curve. On the other hand, in the case of PAAm solution shown in Fig. 1, mixing of the two fluids begin at the $N=2$ curve where the movement of the area with fluorescence in the spanwise direction is observed. In the area further downstream, the two fluid will create multiple layers, and the fluorescence distribution becomes uniform presenting the enhancement in the fluid mixing. Such tendency agrees with the results of Burghelea et al. (2004).

The mixing enhancement observed in the case of PAAm solution is attributed to the unsteady flow that is produced only in the PAAm case and not in the sucrose case. During the visualization carried out at different streamwise positions, several types of behaviors in the fluid mixing were observed. One of them is the ”flapping” motion that is observed mainly in the upstream area of the serpentine channel where the flow begins to be unsteady. Figure 2 shows the flapping motions
Figure 2. Photographs of the instantaneous fluorescence distributions at the $N=2$ curve.

Figure 3. Time variation of the fluorescence distributions, and the mixing index $\phi$ distributions in relation to Reynolds number $Re$.

presented by the instantaneous fluorescence distributions in the time period of $0 \leq t \leq 700\text{ms}$ at the $N=2$ curve. At $t=0\text{ms}$ shown in Fig. 2 (a), a clear boundary of the fluorescence similar to the one observed in the sucrose case is formed. As the time elapse, the area with fluorescence expands outwards in the radial direction, and then move inwards returning to the first position ($t=0\text{ms}$). This motion in the radial direction was observed periodically, and is attributed to the radial flow generated by the viscoelastic fluid which will be discussed shortly.

To evaluate the fluctuating characteristics of the fluorescence distribution in the sucrose and PAAm cases, the time variation of the fluorescence intensity $I_y$ distributions measured in the channel spanwise direction is shown in Figs. 3(a) and (b). The values are measured at the corner of the $N=13$ curve, i.e. $\theta=90^\circ$. $I_y$ is normalized by the spatial average value $I_m$.

The $I_y/I_m$ distribution in the sucrose case shown in Fig. 3(a) remains nearly constant against time $t$, and the areas of larger $I_y$ and smaller $I_y$ are clearly divided at the center of the channel. This shows that the mixing at the boundary is mainly attributed to the molecular diffusion, and there is no secondary flow that is strong enough to enhance the mixing. On the other hand, $I_y/I_m$ distributions in the PAAm case shown in Fig. 3(b) varies as the time $t$ passes presenting a highly unsteady behavior of the fluorescence pattern. Further, $I_y/I_m$ is nearly unity across the channel width and also the time axis, which shows that the fluorescence distribution is uniform and is well mixed.

In order to discuss the influence of the flow Reynolds number $Re$ on the time mean mixing performance of PAAm and sucrose cases at $N=13$, the relationship between the $Re$ and the mixing index $\phi$ is shown in Fig. 3(c). $\phi$ presents the deviation of the fluorescence intensity $I_y$ from the average value $I_m$ and is defined as Eq. (1). $\sigma$ is the standard deviation of $I/I_m$.

$$\phi = 1 - \frac{\sum_{j=1}^{m} \sigma_j}{m} \quad \text{where} \quad \sigma_j = \sqrt{\frac{\sum_{t=1}^{n} (I_{i,j}/I_m,j - 1)^2}{n}}$$

At the smallest $Re$ considered in this study, i.e. $Re = 1.8 \times 10^{-4}$, $\phi$ of the PAAm cases
two cases showing that fluctuation is isotropic. As previously mentioned, the flow is steady in

In the measurement, the fluorescence distributions of the two cases were very similar, namely, the clear boundary of the fluorescence remained steady. As Re increases φ increases markedly, and becomes 0.82 at Re = 6.7 × 10^{-4}. Above this Re, φ then remains constant around 0.82. It is expected that the flow transits from steady to unsteady state in the vicinity of this Re.

In the sucrose solution case, φ becomes maximum at the minimum Re considered in this study, Re = 1.8 × 10^{-1}. φ then decreases as Re increases that is an opposite tendency to the PAAm case. During the measurement although the Re was varied, the boundary position of the fluorescence remained steady and no fluctuation was observed in the visualization measurement. The reason why φ decreases with Re is attributed to the decrease in the Peclet number, that indicates the influence of the molecular diffusion during the flow flows along the serpentine channel decreases as the flow velocity decreases. Comparing the results of sucrose solutions with PAAm case, the enhancement in the mixing performance is obvious. At Re = 3.6 × 10^{-3}, φ in the PAAm case shows 8 times greater value compared with the sucrose case. On the contrary, for smaller Re both values are expected to continuously approach and match with each other.

3.2. Flow velocity measurements

3.2.1. Point measurement and analysis  To first evaluate the fluctuation characteristic of the unsteady flow in the serpentine channel, the velocity variation at a point will be discussed. Figure 4 shows the instantaneous velocity u distributions in the cases of sucrose and PAAm solutions. The measurement position is the channel center at the corner ((r − Rc)/W = 0.5 and θ = 90°) of the N = 2 curve. The subscripts θ and r for each value present the tangential and radial velocity components in the curved area. The Reynolds number is Re = 2.2 × 10^{-3} that is the condition at which the flow is steady in the sucrose case and unsteady in the PAAm case, respectively.

First, the distributions in the sucrose case shown in Figs. 4(a) and (b) contain high frequency fluctuations for both uθ and ur values. The amplitude and frequency are nearly identical in the two cases showing that fluctuation is isotropic. As previously mentioned, the flow is steady in
the sucrose solution case, and the fluctuation is attributed to other reasons. It is, actually, due
to the Brownian motion of the tracer particles that becomes noticeable for the measurement in
the microchannel owing to the scale effect.

In the PAAm case shown in Figs. 4(c) and (d), a fluctuation with frequency and amplitude
equivalent to those obtained in the sucrose case is observed for \( u_\theta \) and \( u_r \) distributions. Therefore,
this is considered to be attributed mainly to the Brownian motion of the particles. On the other
hand, additional fluctuation of relatively low frequency and large amplitude is observed in the
distribution. This fluctuation presents the unsteady behavior of the flow that leads to the
enhancement in the fluid mixing.

The results of the cross spectral analysis carried out for the previously discussed instantaneous
data is shown in Fig. 5. Comparing the results in the cases of sucrose and PAAm solutions, no
peak is observed in the sucrose case and the distribution is flat. In the case of PAAm solution,
however, a peak is clearly observed at the frequency of approximately 0.4Hz, and the fluctuations
of \( u_\theta \) and \( u_r \) are in a coherent state. The two results differ in the area of a frequency smaller
than 5Hz. Above this frequency, the spectrum shows identical distribution indicating that the
high frequency random motion due to the Brownian motion are nearly the same in the sucrose
and PAAm cases. Therefore, the low frequency fluctuation is the major difference in the two
cases.

As shown in Fig. 5(b), the scattering data is uniformly distributed and there is no correlation
between \( \varphi \) and the frequency \( f \) in the sucrose case. On the other hand, in the PAAm case shown
in Fig. (d), the scattering distributions are concentrated in the area adjacent to \( \varphi = 0 \) and 360°
at the low frequency range showing that fluctuating of \( u_\theta \) and \( u_r \) are in phase.

3.3. Cross-sectional distributions
Figure 6 shows the cross-sectional distributions of the streamwise and spanwise components
of the time mean velocities and the fluctuation components \( \bar{u}_\theta \) and \( \bar{u}_r \) measured at the
corner(\( \theta = 90^\circ \)) of the \( N = 2 \) curve. The values with \( \tilde{~} \) are the average data of the adjacent
points for the time period of 0.2s. The fluctuation due to the Brownian motions are considered
to be subtracted by this process and the values present the low frequency fluctuations only.
The influence is particularly large in the streamwise velocity distribution where the distribution becomes more uniform and deviates in the spanwise direction. This is considered to be due to the similar reason explained by Larson et al. (1990) for viscoelastic fluids in Taylor-Couette flows.

These results influence the time mean velocity distributions shown in Figs. 6(a) and (b). The influence is particularly large in the streamwise velocity distribution where the distribution becomes more uniform and deviates in the spanwise direction. This is considered to be due to the large spanwise flow fluctuation that results in a greater fluid transport in the spanwise direction.

Next, comparison is made with the results of the $N = 2$ and 13 curves in order to consider the influences of the streamwise locations in the serpentine channel. Figure 7 shows the cross-spectrum in the $N = 13$ case of $Re = 2.2 \times 10^{-3}$. Figure 8 shows the $\overline{u}$ and $\ddot{u}$ distributions. In the $N = 13$ case, the power increases as the frequency decreases and shows a relatively flat distribution in the range of $0.1 \sim 0.5Hz$. However, the apparent peak observed at $f = 0.4Hz$ in $N = 2$ case cannot be found in $N = 13$ case. Further, the power itself shows a smaller value in $N = 13$ case than in $N = 2$ case showing that the fluctuation amplitude decreases.

In the $\varphi$ distribution, the scattering distribution in the lower frequency range shows a greater dispersion in the $N = 13$ case indicating that the coherence is slightly smaller than in the $N = 2$ case. This tendency can be observed also in Figure 8. Although the $\ddot{u}_\theta$ and $\ddot{u}_r$ are much larger than the sucrose case, both values decreases compared with $N = 2$ case. Further, the difference between $\ddot{u}_\theta$ and $\ddot{u}_r$ in $N = 13$ case becomes smaller. The decrease of the fluctuation amplitudes is considered to influence the time mean distribution also. As shown in Figures 8(a) and (b), not much difference is observed in the $\overline{u}$ distributions compared with the sucrose case. Although not shown here, the fluorescence images taken by the high speed camera indicated that $\Phi$ flapping $\ddot{u}$ motion observed at the $N = 2$ curve does not exists at $N = 13$. This will be discussed in the
mixture of several waves, and actually consists of various vortices and fluctuations. Indeed, in

Figure 7. Cross spectrum in the cases of sucrose and PAAm at the \( N = 13 \) curve.

Figure 8. Mean velocities and fluctuation intensities in the cases of sucrose and PAAm at the \( N = 13 \) curve.

following section in which the two-dimensional flow pattern is discussed.

3.3.1. Two-dimensional velocity distribution Figure 9 shows the contour distributions of the fluctuation components of the streamwise velocity \( \tilde{u}_y \) at the location of \( N = 2 \) curve. The velocity vectors are superposed on the contour map. The flow field in the PAAm case includes low frequency fluctuation and the high frequency random fluctuation due to the Brownian motion. The low frequency fluctuation shows a power peak at the frequency of 0.4 Hz, however, is a mixture of several waves, and actually consists of various vortices and fluctuations. Indeed, in
the two-dimensional velocity distributions measured in this study, several types of flow fields and three-dimensional vortices were observed. Among these, those which represent the typical flow pattern at the period when the velocity increases, decreases, or is equal to the time mean velocity distribution are shown in Fig. 9.

In Fig. 9, one can see that the fluctuations in the flow field is not randomly distributed but shows some tendency. In Fig. (a), an area with positive $\tilde{u}_\theta$ appears and covers a certain area of the channel. In Fig. (b), a wide area with negative $\tilde{u}_\theta$ is observed similarly. It should be noted that sometimes such area covered over the area of one curve unit. Therefore, additional experiment was carried out in the straight part located upstream of the serpentine channel to confirm that no oscillating flow was generated. No such flow was observed, thus it is believed that a large scale three-dimensional vortex is generated in the channel. The area size or the velocity magnitude varied by time and location, and are not completely periodic or regulated. However, such flow field is considered to be the main cause for the flow fluctuation observed in the previous figures.

These kinds of flow fields appear in the channel at the frequency of $0.1 \sim 0.5$Hz. This frequency corresponds to those of the peak observed in the cross spectrum and the flapping motion observed in the visualization measurement. Furthermore, considering the mean streamwise velocity and the area length of this increased/decreased velocity regions, the time period in which this region occupies a curve was approximately 150ms. Since the spanwise velocity is in the order of 0.5mm/s, the fluid will fluctuation in the spanwise length of 75$\mu$m. This corresponds to the channel width and also the variation length of the flapping motion observed in the visualization.

Paying attention on the velocity vectors in the figures, the vectors are directed outwards of the channel curve when $\tilde{u}_\theta$ is positive (increased velocity field) and are directed inwards when $\tilde{u}_\theta$ is negative (decreased velocity field). This is considered to be attributed to the following reason. As often the case in viscoelastic fluids, the polymers (or the solution) is stretched in the streamwise direction due to the shear stress produced by the flow. This provides normal stresses in the spanwise and streamwise direction. Considering then that the streamlines are curved along the serpentine channel, the normal stress particularly produced in the streamwise direction will work in the tangential direction. If a time lag exists in these forces, the forces and the resulting velocity fluctuations will appear with a certain angle to the streamwise direction in combination with the stress in the spanwise direction.

From the other measurement carried out in this study, it is known that these areas were mainly generated in the $N = 1$ and 2 curves. In the downstream area, however, the fluctuations are a mixture of those generated at each streamwise location and those provided from the upstream. The vortices from the upstream will enhance the mixing performance in the curved channel located further downstream of the serpentine channel, however, is expected to also disturb the generation of low frequency fluctuation observed in the $N = 1$ and 2 curves. This is considered to be the main reason why the fluctuation decreases and becomes isothermal in the case of $N = 13$ compared with $N = 2$.

Where and how such large scale fluctuation is precisely generated still remains as a question to be solved. Further measurement and discussion is required to evaluate the vortex structure observed at the curves of the channel. Nevertheless, the present measurement has quantitatively confirmed the velocity fields and provided some insights for understanding the mechanism of the unsteady flows in serpentine channel.

**Conclusions**

Flow characteristics of the viscoelastic fluid flow in the serpentine microchannel were measured and described quantitatively in this study. The fluorescence visualization measurement showed that the mixing performance markedly increased above the Reynolds number of $6.7 \times 10^{-4}$ at
Figure 9. Typical two-dimensional flow patterns presenting the flapping motion at the $N = 2$ curve in the PAAm solution case.

which transition from steady to unsteady flow occurred in the polyacrylamide (PAAm) solution case. The mixing performance enhanced significantly compared with the sucrose solution case. The velocity measurement showed that low frequency flow fluctuation in the spanwise direction of approximately 0.4Hz was generated in the upstream area in the PAAm case. In this area, relatively large areas of high and low velocities were appeared periodically in the curved area. On the other hand, such large scale fluctuation was hardly observed in the downstream area of the channel.

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