Model-free rheometry based on unsteady velocity profile analysis

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With the aim of obtaining a constitutive relation of viscoelastic fluid experimentally, velocity distribution is measured using two kinds of velocimetry; particle tracking velocimetry (PTV) and ultrasonic velocity profiler (UVP). As its configuration suitable for velocimetry-based rheometry, concentric cylinders with a large radius ratio are employed so that spatiotemporal distribution of shear stress in the gap is derived from equation of motion. The results are summarized as correlations among three quantities, i.e. shear rate, strain and stress. PTV is applied for obtaining velocity, and also the constitutive relation as a reference. The operating condition is chosen to involve both elastic and viscous dominant regions in the flow: The flow driven right after the stepwise start of the inner cylinder rotation is dominated by elastic region where stress is generated by strain. The steady state, on the other hand, corresponds to viscous region where shear rate induces stress. Application of UVP for the same purpose is in half way while velocity profile of silicone oil has been obtained. A point of discussion in UVP-rheometry is how measurement noises underlying in the velocity distribution affects on the characterization of fluid.

Keywords: Rheometry, rheology, viscoelasticity, unsteady flows, shear thinning

1 INTRODUCTION

With increasing use of viscoelastic materials in industry, it is required to describe and predict motion of viscoelastic fluids. Motion of Newtonian fluids is described by three equations; equation of motion, continuous equation and Newton's law of viscosity. In cases of viscoelastic fluid, on the other hand, Newton's law of viscosity cannot be employed. Thus in order to describe motion of viscoelastic fluids, alternative equation to Newton's law of viscosity is necessary. The alternative equations, called constitutive equations, have to determine the relationship between fluid deformation and stress generation.

Constitutive equations have been suggested based on microstructure of fluids. Most of them are developed in the field of polymer chemistry. This kind of constitutive equation is typically originates from dynamics of polymer chains. For example, beads connected with springs are introduced as a model to explain the motion of polymer chains, and polymeric fluids are described as an aggregation of them. This kind of equation can describe flow of polymeric liquids correctly, but they are not applicable for viscoelastic fluids with the other microstructures, for example liquid foods and cements.

Velocimetry for fluids have been applied to characterize non-Newtonian fluids. In the case of UVP-PD, velocity profile of the flow through a pipe is measured by ultrasonic velocity profiler (UVP). By combining with pressure difference measurement, the relationship between shear rate and stress is obtained. The advantage of velocimetry-based method is that they are applicable for fluids with any kinds of microstructure. Furthermore, opaque fluids can also be characterized by introducing UVP as velocimetry. UVP-PD succeeded in characterizing many kinds of opaque fluids including foods[1] and cements[2].

One drawback of velocimetry-based characterization is that measurement object has been restricted to steady flows. It is impossible to characterize viscoelasticity from the information about steady flow because flow after the stress relaxation is observed in steady states. One of difficulties to characterize fluids from unsteady flows is to obtain spatiotemporal stress distribution. In steady states, stress at boundary is enough to estimate stress distribution. In unsteady states, on the other hand, equation of motion has to be solved to derive spatiotemporal stress distribution.

The objective of this research is to develop the method to obtain viscoelastic constitutive equation. As velocimetry, particle tracking velocimetry (PTV) and UVP are introduced. Firstly, a constitutive relation of a viscoelastic fluid was obtained from PTV measurement. Through the process to obtain the relation, influences of viscoelasticity on velocity, shear rate and stress distributions are discussed. Secondly, a flow of Newtonian fluid was measured by UVP. From the obtained velocity profile, we discussed how errors on velocity profile affect on the fluid characterization. As a measurement configuration, concentric cylinders with a large gap were chosen. Under the assumption of axial symmetry and two-dimensionality, equation of motion is simplified enough to solve and give spatiotemporal distribution of stress.

2 EXPERIMENT

2.1 Experimental apparatus

Figure 1 shows the experimental apparatus. The configuration is concentric cylinder as Fig. 1 (a)

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indicates. The radius of the inner cylinder r_i and that of outer cylinder r_o are 15 mm and 71.5 mm, respectively. The inner cylinder starts to rotate at t = 0. In this report, the rotational speed is set as 1.02 rps because a viscoelastic behavior is observed clearly in this condition.

In the experiment, velocity profile and rotational torque of the inner cylinder are measured. The velocity profile is obtained by two different measurement techniques, UVP and PTV. The measurement line of UVP is 5.0 mm away from the wall of the inner cylinder to prevent reflection of the ultrasound. A high speed video camera (HSV) is set as shown in Fig. 1 (b), and a horizontal cross section is illuminated with a laser light sheet. The obtained particle images are calibrated by comparing to the grid sheet image obtained before the experiment. The inner cylinder is connected to a torque meter (Visco88, Malvern), and time variation of the rotational torque is recorded.

As test fluids, two kinds of fluid are used. The first one is silicone oil (KF96-1000cs, Shin-Etsu Chemical) which is regarded as a Newtonian fluid whose viscosity is 0.97 Pa·s. The second one is 0.5 wt% polyacrylamide (PAA, AP805C, DiaNitrix) solution which is regarded as a viscoelastic and shear thinning fluid. In the silicone oil and the PAA solution, resin powders, FLO-BEADS CL-2507 (Sumitomo Seika Chemicals), and DIAION HP20SS (Mitsubishi Chemical) are dispersed, respectively, as tracer particles.

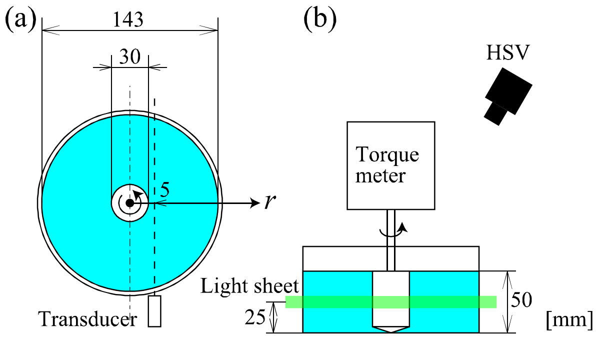

Figure 1: (a) The top view and (b) the side view of the experimental apparatus

2.2 How to characterize viscoelasticity

In order to characterize viscoelastic fluids without any model, the correlations among shear rate, strain and stress should be obtained. In these quantities, shear rate $\dot{\gamma}_{r\theta}$ and strain $\gamma_{r\theta}$ are derived simply from a velocity profile as following equations

$$\dot{\gamma}_{r\theta} = \frac{\partial u_\theta}{\partial r} - \frac{u_\theta}{r}, \qquad (1)$$

$$\gamma_{r\theta}(t) = \int_0^t \dot{\gamma}(\xi) d\xi, \qquad (2)$$

where ξ is an integrating variable which has a dimension of time. In this concentric cylinder configuration, spatiotemporal stress field is derived by solving equation of motion. By assuming homogeneity for tangential and axial direction, and $u_r = u_z = 0$, equation of motion is simplified to

$$\rho \frac{\partial u_\theta}{\partial t} = \frac{2\tau_{r\theta}}{r} + \frac{\partial \tau_{r\theta}}{\partial r}. \qquad (3)$$

The left-hand side of Eq. (3) is derived from spatiotemporal velocity profile. Thus spatiotemporal stress field is obtained by solving Eq. (3) as differential equation. In order to solve this, boundary condition is necessary. Then stress at the wall of the inner cylinder τ_{wall} is obtained from the torque to rotate the inner cylinder T. The relationship between τ_{wall} and T is described as

$$\tau_{wall} = \frac{T}{2\pi r_i^2 h} \qquad (4)$$

where h is the height of the inner cylinder (50 mm). By the process shown above, spatiotemporal distributions of shear rate, strain and stress are obtained. Thus local viscosity is defined as the ratio of stress to shear rate:

$$\eta = \frac{\tau_{r\theta}}{\dot{\gamma}_{r\theta}}. \qquad (5)$$

Another way to represent property of fluids is direct plot of the correlations among shear rate, strain and stress like Fig. 4. The way to interpret the plot is explained in the Section 3.2.

3 EXPERIMENTAL RESULTS

3.1 Characterization of Newtonian fluid with PTV

In the first experiment, velocity field of the silicone oil is measured by PTV. Shear rate, stress and viscosity are derived from the velocity field. The aim of this experiment is a demonstration of stress and viscosity distributions with Newtonian fluid whose viscosity is known. Figure 2 (a) shows the velocity profile. The velocity is normalized by the velocity of the inner cylinder wall U_{wall} = 96 mm/s. Right after the inner cylinder starts the rotation at t = 0, fluid near the inner cylinder is driven. After that velocity diffuses to outer region as time advances. Figure 2 (b) shows the shear rate distribution derived from Fig. 2 (a) with Eq. (1). The shear rate is basically negative because of the definition of the spatial axis. Theoretically, the magnitude of shear rate is large near the inner cylinder, and decreases along r axis. The steady state in Fig. 2 (b) basically follows this tendency, but deviates from the theory at around r = 20 mm and 65 mm. In these regions, it is difficult to measure velocity correctly because of light reflection from the inner and outer cylinder wall. Furthermore, measurement error on the velocity is enhanced in Eq. (1) because the differential is calculated as finite differential. Figure 2 (c) indicates stress distribution obtained by solving Eq. (3). Stress in Fig. 2 (c) is also basically negative because of the coordinate definition. In the steady condition, stress distribution is described theoretically as

$$\tau_{r\theta} = \left(\frac{r_i}{r}\right)^2 \tau_{wall}. \qquad (6)$$

The stress distribution in Fig. 2 (c) almost follows Eq. (6) in the steady state. Thus viscosity is estimated properly in the steady state in Fig. 2 (d), especially around at 25 mm < r < 45 mm, t > 2.0 sec: In this region, the obtained viscosity is about 1 Pa·s while the reference value is 0.97 Pa·s. Near the inner and the outer cylinder wall, viscosity value in Fig. 2 (d) extremely differs from the reference value. This kind of error is caused by the error enhancement in the shear rate calculation.

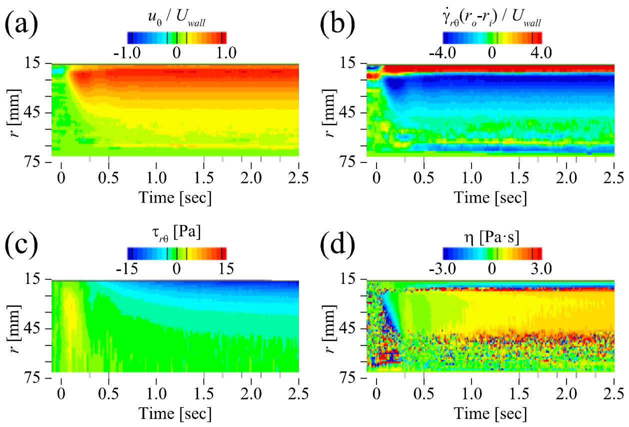

Figure 2: Spatiotemporal distribution of (a) tangential velocity (b) shear rate (c) shear stress and (d) viscosity of 0.97 Pa·s silicone oil obtained by PTV

3.2 Characterization of viscoelastic fluid with PTV

The four quantities in Fig. 2, velocity, shear rate, stress and viscosity, are obtained also for the PAA solution as shown in Fig. 3. The aim of this measurement is to show how viscoelasticity appears in these quantity distributions. In contrast to the Newtonian case in Fig. 2 (a), velocity decreases as time advances after t = 0.5 sec. At this time, velocity has local minimum at around r = 35 mm. After that velocity is at minimum at around t = 1.5 sec in wide range of r, and accelerated again. This velocity oscillation is one of the characteristics of the viscoelastic fluid flow. In the steady state after t = 2.0, the fluid only near the inner cylinder moves compared to the Newtonian case in Fig. 2 (a). This is caused by shear thinning: Viscosity becomes lower near the inner cylinder because of high shear rate. Viscoelasticity appears in the stress distribution, Fig. 3 (c), as the local minimum of $|\tau_{wall}|$ at around t = 1.5 sec. In the steady state, stress distribution follows Eq. (6) which holds even in non-Newtonian flows. Figure 3 (d) indicates that viscoelasticity is expressed as negative viscosity if local viscosity is defined by Eq. (5). At around t = 0.5, when viscoelasticity explicitly affect to fluid motion, shear rate takes positive value as Fig. 3 (b) indicates while stress takes positive value. Thus locally-defined viscosity takes negative value as a reflection of the viscoelastic behavior.

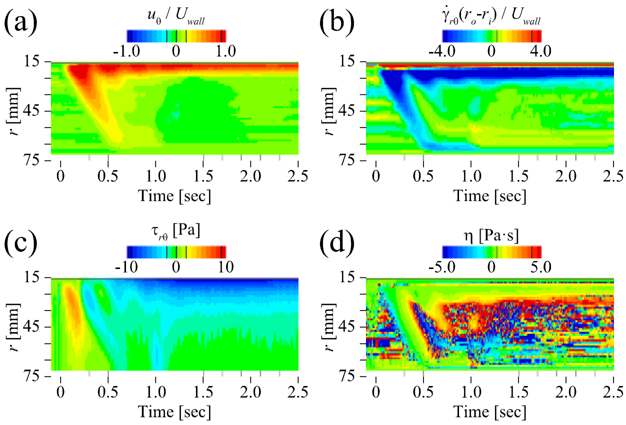

Figure 3: Spatiotemporal distribution of (a) tangential velocity (b) shear rate (c) shear stress and (d) viscosity of 0.5wt% PAA solution obtained by PTV

As a way to display viscoelastic property, we suggest a plot which shows the correlations among shear rate, strain and stress. The quantity fields in Fig. 3 are converted to this plot as shown in Fig. 4. The horizontal axis, vertical axis and color represent shear rate, strain and stress, respectively. In this plot, elastic property is expressed as strain-dependent stress. In Figure 4, stress depends on the strain in the region of $\gamma_{r\theta}$ < 5.0. If strain exceeds 5.0, on the other hand, stress is independent on strain. Instead, shear rate is the main factor to decide stress. It means that the PAA solution is viscous state in this region. The PAA solution is switched elastic regime to viscous regime at around $\gamma_{r\theta}$ = 5.0. Information about viscoelastic property appears also as the shape of lines in Fig. 4. The wavy shape of lines means that the value of shear rate is fluctuating periodically at each spatial position. This corresponds to the oscillatory motion of the fluid which is caused by viscoelasticity. The frequency of the oscillation should be determined by the relaxation time of the PAA solution.

The relationship plotted on Fig. 4 is constitutive relation itself. If this relation is employable as alternative of constitutive models in numerical simulations, the concentric cylinder system works as a generator of constitutive relations. From a practical point of view, however, non-homogeneity of data points on shear rate axis is regarded as a

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problem when Fig. 4 is employed as constitutive relation. The absence of data points in high shear rate region is attributed to the fact that data points distributes on r axis homogeneously although high shear rate appears only in the thin region near the inner cylinder. Therefore, it is important to obtain velocity distribution which is highly-resolved near the inner cylinder, or interpolate velocity properly by introducing function approximation.

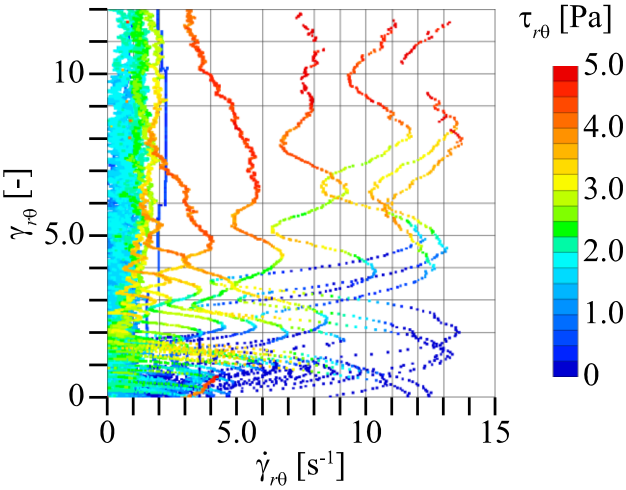

Figure 4: The correlations among shear rate (horizontal axis), strain (vertical axis) and stress (color) of 0.5wt% PAA solution obtained by PTV

3.3 Characterization of Newtonian fluid with UVP

With the aim of clarifying difficulties to construct the fluid characterization by UVP, flow of the silicone oil is measured by UVP. The velocity profile in Fig. 5 (a) is obtained as a result of simultaneous measurement of HSV and UVP. The tangential velocity component u_θ is derived from the projected velocity component on the measurement line u_{UVP} by the following equation

$$u_\theta = \frac{r}{d} u_{UVP} \qquad (7)$$

where d is the offset of the measurement line from the center of the cylinder (d = 20 mm). Equation (7) holds under the assumption of $u_r = 0$. Velocity profile cannot be obtained in the region of r < 20 mm because of the offset. Thus stress at r = 20 mm is referred from Fig. 2 (c) as a boundary condition to obtain Fig. 5 (c).

According to Fig. 5 (a), it is difficult also for UVP to obtain velocity near the inner cylinder. In the steady state, Fig. 5 (a) follows the theoretical velocity distribution only in the region of r > 25 mm, and largely deviates in r < 25 mm. It is expected that UVP cannot measure correct velocity profile because of the dispersion of the ultrasound beam. This measurement error is enhanced by the differential calculation in Eq. (1), and result in the extreme viscosity values in Fig. 5 (d). Fortunately, proper viscosity value is obtained at around r = 30 mm in the steady state of Fig. 5 (d). This fact implies that properties of fluids are properly derived if shear rate is obtained with less noise. Thus we are constructing function approximation for spatial velocity distribution with the aim of noise reduction on the shear rate, and interpolation of velocity near the inner cylinder.

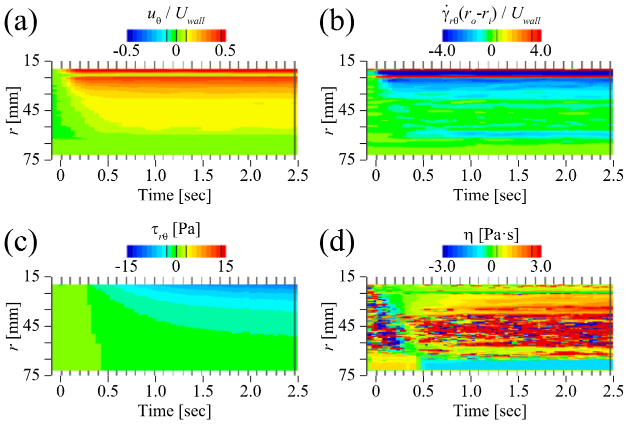

Figure 5: Spatiotemporal distribution of (a) tangential velocity (b) shear rate (c) shear stress and (d) viscosity of 0.97 Pa·s silicone oil obtained by UVP

4 CONCLUSION

Velocity profile between the concentric cylinders is measured with the aim of obtaining constitutive equation experimentally. Spatiotemporal distributions of stress and local viscosity are derived by substituting the velocity profile into equation of motion.

In the PTV measurement for viscoelastic PAA solution, elasticity appears as negative local viscosity. By plotting stress as a function of shear rate and stress, the switching from elastic regime to viscous regime is expressed. The function is a constitutive equation of the test fluid which is obtained experimentally without introducing any rheological model.

In order to construct this viscoelastic characterization based on UVP, error enhancement in the shear rate calculation is a problem to solve. The obtained property such as local viscosity differs from the reference value because of the error enhancement. We are planning to solve this problem by introducing polynomial function approximation for velocity profiles.

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