Tubeless Syphon Flow of Viscoelastic Suspensions

A tubeless syphon has been employed by various authors¹⁻³ to investigate the extensional viscosity of polymer solutions since the efficacy of this technique for the extensional viscosity measurement was rediscovered by Astarita and Nicodemo.⁴ In the tubeless syphon apparatus, the sample liquid is continuously sucked into a nozzle which is above the free level of liquid. The technique of forming a vertical free boundary column of liquid looks attractive because the liquid is in its quiescent state until the extensional flow begins, whereas, in the spinning configuration, the liquid experiences certain shear history prior to exit from the orifice. However, there are inherent difficulties in using the tubeless syphon. Since the column profile depends on the flow properties of liquid, the extensional rate is not necessarily constant along the column. Therefore, the flow fields are unsteady in the sense that the normal stress applied to a fluid element and the resultant extensional rate are transient.

In the present note, the transient extensional viscosity as a function of strain is obtained for a polystyrene solution through the procedure described before.^{5,6} The effect of addition of silica particles on the extensional viscosity is discussed.

EXPERIMENTAL

The principle and the apparatus of tubeless syphon technique has been reported in the previous paper.⁵ When a stable flow is obtained between the nozzle tip and the free level of liquid at a given flow rate Q, the force exerted by the liquid on the nozzle entrance is directly measured and a photograph of the syphon is taken. The normal stress σ as a function of distance x along the column is calculated from the geometrical function.

The sample used were a solution of polystyrene ($M_w = 8.8 \times 10^6$) in diethyl phthalate at a concentration of 3 wt % and a silica suspension in the polystyrene solution at a concentration of 1 wt %. The silica was Aerosil 300 (Degussa Co.). After mixing by hand, the suspension was stored for 1 day and was sufficiently degassed before measurements. The experiments were carried out at 25°C.

The viscosity behavior in shear flow was first tested by a coaxial cylinder rheometer. Figure 1 shows the shear rate dependence of apparent viscosity for the polystyrene solution and the suspension. The apparent viscosity is constant at low shear rates and decreases with increasing shear rate. The Newtonian viscosity of the suspension is approximately 9 times that of the medium.

RESULTS AND DISCUSSION

Figure 2 shows the dependence of velocity v on the position x along the column at a flow rate of 0.119 cm·s⁻¹ for the polystyrene solution. The extensional rate G (= dv/dx) is not constant along the flow direction, but increases with position x. It has been reported^{1,5} that, in the case of polyacrylamide solutions, the velocity linearly increases and the extensional rate is almost constant. The column profile probably varies depending on the relaxation time of liquid. Thus it must be stressed that the tubeless syphon method is noncontrollable. The variable extensional rate makes it difficult to understand the flow behavior.

In the tubeless syphon experiments, the flow fields are unsteady in the Lagrangian sense. The normal stress varies with time or duration of deformation of a given fluid element even though the extensional rate is constant. Figure 3 shows the time dependence of normal stress at different flow rates for the polystyrene solution. The time t necessary to a fluid element to go from the free level to the position x is calculated by the following equation:

$$t = \int_0^x \frac{dx}{v} \tag{1}$$

The normal stress rapidly increases with time at higher flow rates, but its value ranges from 10^1 to 2×10^3 Pa, irrespective of flow rate. Since the velocity increases with increasing flow rate, it seems likely that the time scale is chiefly affected by flow rate.

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Fig. 1. Shear rate dependence of apparent viscosity for polystyrene solution and silica suspension: (0) medium; (\bullet) 1 wt % suspension.

The extensional flow behavior should be discussed in terms of total strain and extensional viscosity. The strain ϵ , which is experienced by a fluid element since the onset of flow, and the extensional viscosity η_{el} are defined as follows:

$$\epsilon = 2 \ln[R(0)/R(x)] \tag{2}$$

$$\eta_{\rm el} = \frac{\sigma}{G(x)} = \frac{\sigma}{dv/dx} \tag{3}$$

where R(x) is the syphon radius as a function of distance x along the column.



Fig. 2. Dependence of velocity v on position x along the column at a flow rate of 0.119 cm³·s⁻¹ for polystyrene solution.



Fig. 3. Dependence of normal stress σ on time t at different flow rates for polystyrene solution. Q (cm³·s⁻¹): (O) 0.027; ($\mathbf{\Phi}$) 0.043; ($\mathbf{\Phi}$) 0.082; ($\mathbf{\Phi}$) 0.119.

The extensional viscosity result for the polystyrene solution is shown in Figure 4. All the plots lie very close to a single curve. Although the polystyrene solution used does not show the linear dependence of strain on the time, the extensional viscosity as a function of strain can be obtained.

The analysis was repeated for 1 wt % suspension and the master curve is shown by a broken line in Figure 4. The extensional viscosity increases with strain and the two curves seem to be superimposed by a vertical shift. The most important point with regard to the behavior of suspension is that the presence of silica particles in the polymer solution leads to a decrease of the extensional viscosity while the shear viscosity is increased. The increase in shear viscosity has been explained by the hydrodynamic effect and the existence of flocculates. However, direct applications of these mechanisms to the extensional viscosity are not appropriate because there are significant differences between the shear viscosity and the extensional viscosity. The shear viscosity is a manifestation of the dissipation of energy, whereas the extensional viscosity may be observed as a combined response of viscous and elastic effects. In this study, the extensional rate is in the range of $1-3 s^{-1}$. At these extensional rates, the elastic effect may play a much more dominant role. Elastic response of the polymer solution is attributed to the orientation and extension of the polymer chain in the flow direction. The dispersed particles or flocculates hardly accumulate the energy because they more



Fig. 4. Extensional viscosity as a function of strain for polystyrene solution and silica suspension: (O) medium; (--) 1 wt % suspension.

or less behave like hard units. Therefore, it is considered that the reduction of volume occupied by the polymer solution is responsible for the decrease of extensional viscosity. In addition, configurational rearrangement of flow units occurs during the extension of sample. The rearrangement may lead to shear deformation of liquid between flow units. In the case of polymer solution, the molecular flow can be regarded as a pure extension. But in the presence of particles, the liquid medium is subjected to the extension and shear deformation during flow. Presumably, a prevailing shear deformation between flow units also causes the decrease of extensional viscosity of suspension. However, to establish explanation attempted, detailed experiments will be required.

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YASUFUMI OTSUBO •

Department of Image Science and Technology Faculty of Engineering Chiba University 1-33, Yayoi-cho, Chiba-shi, 260 Japan

KAORU UMEYA

Department of Applied Chemistry Faculty of Engineering Tohoku University Aoba, Aramaki, Sendai-shi, 980 Japan

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* Author to whom correspondence should be addressed.