

Experiments on the Reverse Squeezing Flow of Dilute Polymer Solutions*

(Fluids with Newtonian Viscosity)

Takatsune NARUMI**, Yoshihiro HOSOKAWA***
and Tomiichi HASEGAWA**

The reverse squeezing flow of dilute polymer solutions has been studied by utilizing a flat plate and a spherical surface of a large radius of curvature. In this experiment, the two surfaces are forced to separate by the application of constant forces from a stationary and contacting state. All the liquids used are Newtonian in viscosity. The following points are found by comparing the results of inelastic liquids and viscoelastic liquid. In the case of the viscoelastic liquid, the surfaces separate from each other much more slowly than in the case of the inelastic liquid. This trend is clearly shown when the separation is small. The results predicted by the theory of inelastic fluids with the assumption of the quasi-steady state agree well with the experimental results. The same assumption, however, cannot explain the slow separation effect because of a small contribution of elastic terms.

Key Words: Viscoelastic Fluid, Non-newtonian Fluid, Unsteady Flow, Reverse Squeezing Flow, Squeezing Flow, Dilute Polymer Solution, Experimental Study

1. Introduction

Studies on the flow between two surfaces approaching each other, i. e., squeezing flow, are important in industries relating to lubrication. In particular, there is a possibility of improving lubrication when viscoelastic fluids are utilized as the squeezing film. Also it is of interest to analyze this flow theoretically with examination of the constitutive equation, because squeezing flow involves elongational, shear and unsteady flow characteristics. From these points of view, many experimental and theoretical studies had been conducted on the squeezing flow of viscoelastic fluids^{(1)~(3)}. However, most of these treated normal squeezing flow, that is, that which occurs when two parallel plates approach each other, and of nondilute polymer solutions. One of the examples of this kind is the study by Brindley, Davies and Walters⁽³⁾. The main conclusions of their study were:

(1) The behavior of elastic liquids in a squeezing film situation is predictable from a knowledge of the shear-rate-dependent viscosity only, under appropriate conditions associated with light loading.

(2) Viscoelastic effects are in evidence in squeeze film situations when polymer solutions are exposed to conditions of heavy loading. The solutions behave as better lubricants than would be expected from viscosity considerations.

(3) Under some severe loading conditions, solidlike bouncing behavior is possible.

In an attempt to provide qualitative theoretical predictions of the observed behavior, many rheologists have proposed models. Unfortunately, however, most of them led to conclusions that were the opposite of the result described in (2) above. A clear theoretical explanation for these experimental results has not been provided yet.

On the other hand, in the context of total lubrication, it is important to consider the flow between two surfaces which are separating from each other, because such a flow inevitably occurs after the approaching motion. However, only a few studies, for example, those by Oliver and Shahidullah⁽⁴⁾ and Avila and

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** Faculty of Engineering, Niigata University, 8050 2-no-cho Ikarashi, Niigata, 950-21, Japan

*** Canon Co.,Ltd., 7-5-1 Hakusan, Toride, 302, Japan

Binding⁽⁵⁾, have been carried out on this reverse squeezing flow. Avila and Binding have shown that the separation of the plates in the polymer solutions is slower than that predicted by inelastic theory in the long time scale, except for the rapid separation at the beginning of motion.

In the present study, we are concerned with the reverse squeezing problem, which has not been reported on much either theoretically or experimentally, and try to clarify the behavior of the polymer solutions experimentally. We investigate a case in which two surfaces, a flat plate and a spherical surface of large radius of curvature, are forced to separate by the application of constant forces from a stationary and contacting state.

Water and glycerine solutions were tested as the Newtonian liquids, and PEO solutions (100 ppm and 200 ppm) were utilized as dilute polymer solutions. It is known that effects of elasticity are not expected for solutions of this order of polymer concentration in ordinary squeezing flow⁽⁶⁾. It is unknown, however, whether the elasticity of the above-mentioned polymer solutions is effective or not in the reverse squeezing flow. In the present experiment, we can make a direct comparison between the results for polymer solutions and those for inelastic liquids, because these dilute polymer solutions have the property of Newtonian viscosity.

2. Apparatus and Liquids Used

2.1 Apparatus

A schematic diagram of the apparatus is given in Fig. 1. An optical flat glass (A) and a convex lens (B), both 40 mm in diameter, were set up so as to permit a reverse squeezing flow system. The flat glass was mounted in an acrylic reservoir (C), as is shown in Fig. 1. Three convex lenses, which were 1.00 m, 2.00 m and 3.00 m in radius of curvature, were used in this experiment. A shaft supported by a liner motion bearing (E) was attached to the lens via a holder in order to allow vertical motion of the lens. The top of the shaft was connected to a spring (F), which was adjustable in length by a stepping linear motor unit (G) and displacement transducer (D'). Before the separating motion, the convex lens was kept in contact with the glass plate with an applied downward force by a modified balance (H). In order to measure both the clearance between the two surfaces at the center of plate (we call it the center clearance for short) and the forces transmitted to the glass plate via the liquid film, we attached a displacement transducer (D) and load cells (I), which were connected to a personal computer. The flat glass and the convex lens were lit from below by He-Ne laser parallel beams

through the optical system, so that observation of Newton's rings (optical interference fringes), and hence evaluation of very small clearances and contacting states would be possible.

In the first stage of the experiment, the lens and plate were set under a contacting condition with a force applied by the modified balance, and then adjustment of the contacting state and position was made by the observation of the Newton's rings, such that the contact point was situated at the center of the plate. The contacting force (about 0.5 N downward) and the separating forces (about 0.3 to 1.2 N upward, which were applied when the contacting force was removed) were prescribed by regulation of the spring and the balance. After the system was held at rest, rapid removal of the modified balance caused a separating motion. The transmitted forces and the center clearances were measured with the load cell and the displacement transducer. The very small clearances at the beginning of motion was estimated by analysis of a picture of the Newton's rings, which was filmed by a high-speed camera.

2.2 Liquids used

Five kinds of liquids were tested in this experiment: distilled water (0.957 mPa·s); 14% (1.41 mPa·s) and 30% (2.33 mPa·s) water-glycerine solutions; and 100 ppm (1.16 mPa·s) and 200 ppm (1.37 mPa·s) water-PEO solutions. PEO is a polyethylene oxide of grade number 18 (PEO 18) manufactured by SEITETSU KAGAKU in Japan. All liquids are defined on a weight basis. Tests were performed at $21 \pm 1^\circ\text{C}$. It was

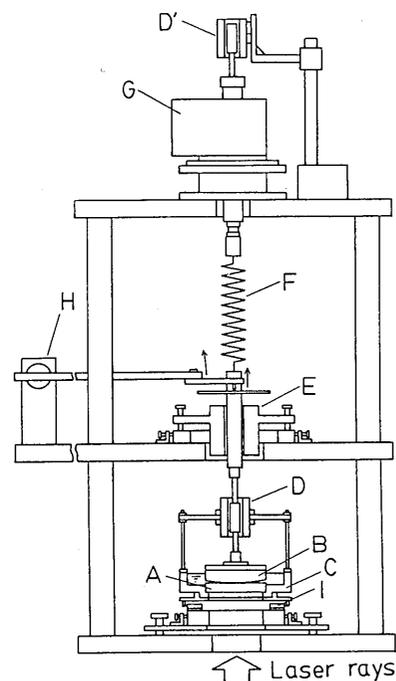


Fig. 1 Apparatus

confirmed by means of a capillary viscometer that all of the liquids used were Newtonian in viscosity (as shown in the above parentheses). In consideration of polymer fracture in high shear flows, fresh PEO solutions were used on each occasion in this test.

3. Results and Discussion

3.1 Analysis for Newtonian liquids

An analysis for inelastic liquids will be performed in the following for comparison of the experimental results. Cylindrical coordinates and factors (a , R , h_a , $h_0(t)$ and h), as shown in Fig. 2, are used for this analysis, and the relations of these factors are given below the figure. The $h_0(t)$, denoting a function of time, will henceforth be abbreviated as h_0 . As assumed in the ordinary lubrication problem⁽⁷⁾, the flow is considered to be inertialess, axisymmetric and quasi-steady because of the slow movement of the lens and the significantly small clearance. Under these assumptions, the r and z components of the equation of motion are

$$0 = \frac{\partial S_{rr}}{\partial r} + \frac{\partial S_{zr}}{\partial z} + \frac{S_{rr} - S_{\theta\theta}}{r} \quad (1)$$

$$0 = \frac{\partial S_{rz}}{\partial r} + \frac{\partial S_{zz}}{\partial z} + \frac{S_{rz}}{r}, \quad (2)$$

where S_{rz} , etc., are the physical components of the total stress tensor. Velocity components are considered as

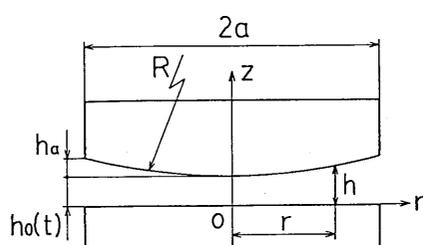
$$v_r = v_r(r, z), \quad v_\theta = 0, \quad v_z = v_z(r, z),$$

so that the equation of motion for Newtonian fluids reduces to the following from the consideration of the order of each term (that is, lubrication assumption) when $r \gg z$.

$$0 = -\frac{\partial p}{\partial r} + \eta \frac{\partial^2 v_r}{\partial z^2} \quad (3)$$

$$0 = -\frac{\partial p}{\partial z}, \quad (4)$$

where p is the isotropic pressure, and η is the viscosity. Since $v_r \gg v_z$, these equations are the same as those derived when the v_z was omitted a priori. Integration of Eq. (3) with respect to z under the bound-



$$h_a = \frac{a^2}{2R}, \quad h = h_0(t) + \frac{r^2}{2R}, \quad \dot{h}_0 = \frac{dh_0}{dt}$$

Fig. 2 Coordinates and nomenclature

ary condition gives v_r as follows.

$$v_r = \frac{1}{2\eta} \frac{\partial p}{\partial r} (z^2 - hz) \quad (5)$$

The continuity equation is integrated to give

$$2\pi r \int_0^h v_r dz = -\pi r^2 \dot{h}_0, \quad (6)$$

and substitution of Eq. (5) yields

$$\frac{dp}{dr} = \frac{6\eta r \dot{h}_0}{h^3}. \quad (7)$$

Although the same result could have been given simply from the Reynolds lubrication equation⁽⁸⁾, the derivation from the equation of motion has been performed in this paper, because the same approach will be useful to analyze the viscoelastic fluids in a later section. Integration of this equation under the boundary conditions of pressure, $p=0$ (atmospheric pressure) at $r=a$ and $h=h_0+h_a$, gives

$$p = -3\eta R \dot{h}_0 \left\{ \frac{1}{h^2} - \frac{1}{(h_0+h_a)^2} \right\} \quad (8)$$

Since the extra stress component τ_{zz} is zero at the surface of the plate and negligible everywhere in the film, the force transmitted to the plate or the lens through the liquid film is given as follows.

$$F = -\frac{3\pi\eta a^4 \dot{h}_0}{2h_a^2} \left\{ \frac{h_a}{(h_0+h_a)^2} - \frac{1}{h_0} + \frac{1}{h_0+h_a} \right\} \quad (9)$$

Considering that $h_0 = h_a/2$ at $t = t_m$, we have the following expression for h_0 by integration of \dot{h}_0 in Eq. (9).

$$\frac{2h_a^2 F}{3\pi\eta a^4} (t - t_m) = \frac{h_a}{h_0+h_a} + \log \frac{3h_0}{h_0+h_a} - \frac{2}{3} \quad (10)$$

3.2 Experimental results

Figure 3 shows typical data measured with the displacement transducer and the load cell. In this figure, the transmitted force (left axis of the ordinates) and the center clearance (right axis) are plotted as a function of time. Tensile forces are

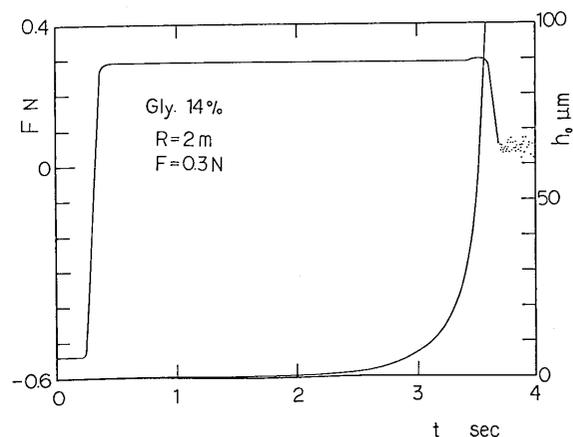


Fig. 3 Transmitted force and center clearance against time

denoted by positive values. It is obvious that the transmitted force took a constant value as soon as the tensile force was applied, and remained constant during the measurement of the center clearance. Since this trend was the same for all cases, we consider the experiment to have been conducted under a constant tensile force.

Next, we will consider the relationship between the center clearance and the time taken from the onset of the motion. Typical results of the glycerine 14% solution and PEO 200 ppm solution, whose viscosities are nearly the same, are presented in Figs. 4 and 5 for the case of $R=2.00$ m and $F=0.6$ N. The data obtained by the optical method are plotted with closed circles and those obtained by the transducer are plot-

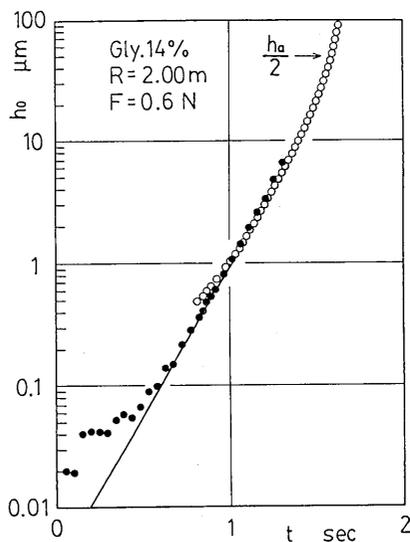


Fig. 4 Center clearance versus time for the glycerine 14% solution

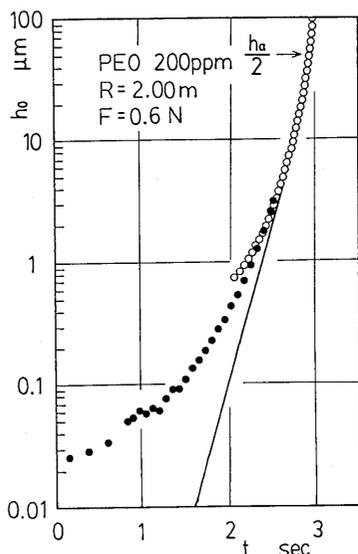


Fig. 5 Center clearance versus time for the PEO 200 ppm solution

ted with open circles. The solid lines are theoretical curves based on Eq.(10), which is drawn so as to coincide with the experimental result at the position where $h_0 = h_a/2$ (the position is shown by arrows in the figures). In the case of glycerine 14% solution (Fig. 4), experimental results are in good agreement with theoretical ones when $h_0 > 0.1 \mu\text{m}$. The data obtained by means of the displacement transducer include rather large errors when $h_0 < 1 \mu\text{m}$, because there are limits in rigidity of the transducer mounting used. The data with significant errors will be omitted in the later discussion. Although it has nearly the same viscosity, the PEO 200 ppm solution (Fig. 5) shows disagreement with the experimental and Newtonian theoretical results except where $h_0 = h_a/2$. Furthermore, the time required for separation is longer for the PEO 200 ppm solution than for the glycerine 14% solution.

3.3 Consideration by means of normalization

Since the comparison between the experimental data and Eq.(10) made in the preceding section lacks generality, we will adopt the following normalized factors.

$$p^* = \frac{p \cdot \pi a^2}{F}, r^* = \frac{r}{a}, h_0^* = \frac{h_0}{h_a}$$

Then, a nondimensional expression of \dot{h}_0 is derived by the normalization of Eq.(7) as

$$\dot{h}_0^* = 6 \cdot \frac{\eta \cdot \pi a^2}{F \cdot h_a} \cdot \frac{a^2}{h_a^2} \cdot \dot{h}_0$$

Further, we obtain normalized Eq.(9) as follows.

$$\dot{h}_0^* = -4 / \left\{ \frac{1}{(h_0^* + 1)^2} - \frac{1}{h_0^*} + \frac{1}{h_0^* + 1} \right\} \quad (11)$$

The generalized relationships between the separ-

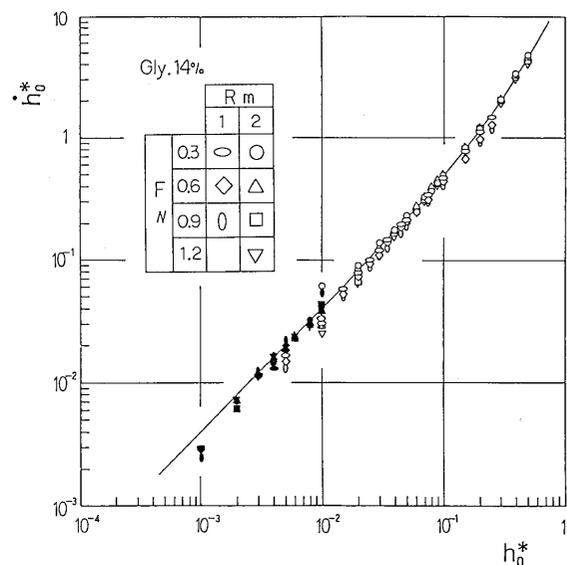


Fig. 6 Normalized relationships between the rates of separation and the center clearance for the glycerine 14% solution

ating rate evaluated by the experimental data and the center clearance are shown in Figs. 6 to 9, where the ordinate is \dot{h}_0^* and the abscissa is h_0^* . Symbols (\circ \square \triangle ∇) presented in the figures denote the data from the transducer, and reversed symbols (\bullet \blacksquare \blacktriangle \blacktriangledown) show the data obtained by the optical method. Equation (11) is also shown by a solid line. In the case of glycerine solutions, the experimental results agree well with Eq.(11). Hence the assumptions made in section 3.1 are regarded as reasonable. The same results are obtained for the distilled water (not shown in this paper). On the other hand, as shown in Figs. 8 and 9, experimental results for PEO solutions show a smaller rate (\dot{h}_0^*) than that predicted by Eq.(11) when the separation is small. This difference diminishes with increase of h_0^* . By inspection of Figs. 8 and 9, it is obvious that these results are independent of separating forces and radii of curvature, but affected by the concentration of solutions (i.e., the elasticity of solutions).

3.4 Contribution of elasticity

In this section, we treat the contribution of elastic terms in the constitutive equation of viscoelastic fluid under the same assumptions used for Newtonian cases. The Denn Model (B), explicit in stress, is utilized for simplicity. It is written as⁽⁹⁾

$$\tau^{ij} = 2m\{4|II_e|\}^{(n-1)/2} \cdot e_{ij} - 2m\lambda\{4|II_e|\}^{(s-1)/2} \cdot \frac{\delta e_{ij}}{\delta t}, \tag{12}$$

where e_{ij} is a component of the rate of strain tensor, $|II_e|$ is the second invariant of the rate of strain tensor, $\delta/\delta t$ denotes the convected derivative (oldroyd derivative), and n, m, λ, s , are the material constants. Since

$n=1$ for all solutions used in this experiment, m becomes η (viscosity in the usual sense). Therefore, Eq.(12) is equivalent to the second-order fluid when the coefficient of the second normal stress difference is omitted. Also, we shall concern ourselves with the case of $s=2$ for the simplification, which is similar to that of the PEO 200ppm solutions where s is about 1.8⁽¹⁰⁾. We now solve the Eq.(12) under the above-mentioned conditions and obtain the following expression for the r component of the equation of motion.

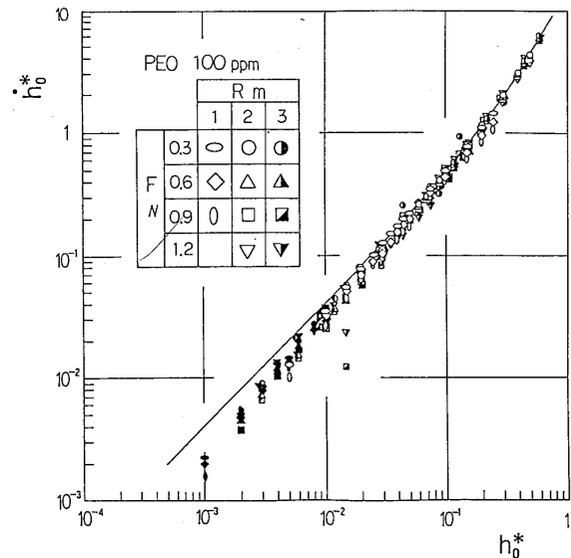


Fig. 8 Normalized relationships between the rates of separation and the center clearance for the PEO 100 ppm solution

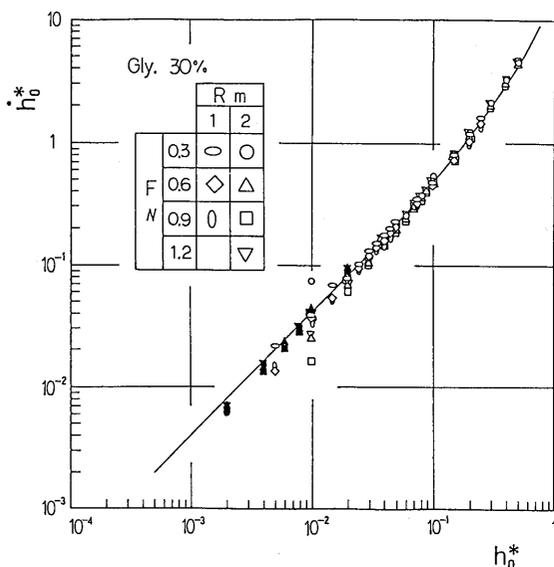


Fig. 7 Normalized relationships between the rates of separation and the center clearance for the glycerine 30% solution

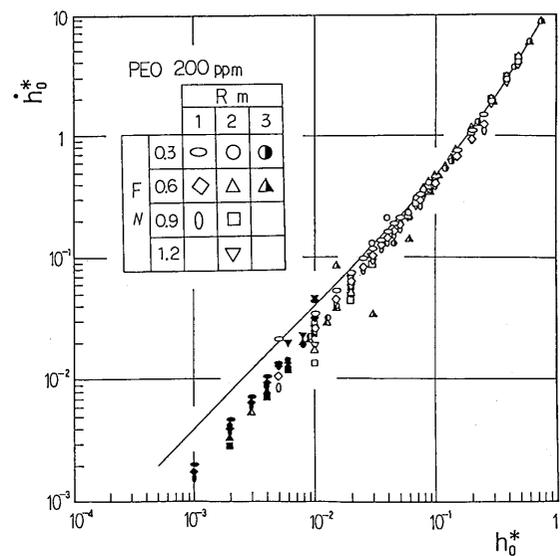


Fig. 9 Normalized relationships between the rates of separation and the center clearance for the PEO 200 ppm solution

$$0 = -\frac{\partial p}{\partial r} + \eta \frac{\partial^2 v_r}{\partial z^2} + \eta \lambda \left\{ 4 \frac{\partial v_r}{\partial z} \cdot \frac{\partial^2 v_r}{\partial r \partial z} - v_r \cdot \frac{\partial^3 v_r}{\partial r \partial z^2} - v_z \cdot \frac{\partial^3 v_r}{\partial z^3} + 2 \frac{\partial^2 v_r}{\partial z^2} \cdot \frac{\partial v_z}{\partial z} + \frac{\partial v_r}{\partial r} \cdot \frac{\partial^2 v_r}{\partial z^2} - 3 \frac{\partial v_r}{\partial z} \cdot \frac{\partial^2 v_z}{\partial z^2} + \frac{2}{r} \cdot \left(\frac{\partial v_r}{\partial z} \right)^2 \right\} \quad (13)$$

Next we consider the order of each term of the above equation. That is, if the order of v_r is written as V and the orders of r and z are equal to those of a and h_a , respectively, we have

$$v_r = O(V), \quad v_z = O(\dot{h}_0) = O(V \cdot h_a/a),$$

where $a \gg h_a$. Since λ is about 10^{-2} to 10^{-3} (sec) for the PEO solutions used in this experiment⁽¹⁰⁾, it is reasonable to regard it as follows.

$$\lambda = O(h_a/v_r) = O(h_a/V)$$

Consequently, the viscosity term becomes $O(V/h_a^2)$ and the elastic terms in which λ is involved become $O(V/(a \cdot h_a))$, where the viscosity η is common to both terms and excluded from the above estimation. In other words, the contribution of the elastic terms is smaller than that of the viscosity term by $O(h_a/a)$. The terms of $O(V/a^2)$ and of smaller magnitude have already been omitted in the derivation of Eq.(13). In the case of Newtonian analysis, we obtained Eq.(4), i.e., $\partial p/\partial z = 0$, from equation of motion (2) under the lubrication assumption. This means that the terms of the same order as the elastic terms were already omitted. For example,

$$\frac{1}{r} \cdot \frac{\partial v_r}{\partial z} = O\left(\frac{V}{a \cdot h_a}\right).$$

In view of the fact that the experimental data of Newtonian fluids were in good agreement with the theoretical result, the elastic terms in Eq.(13) must be negligibly small. In fact, when we solved this equation of motion by perturbation of λ , assuming that the velocity profile would be the same as that of the flow in the parallel plates of the same clearance at any r position, we obtained a solution nearly equal to the Newtonian one⁽¹¹⁾.

The following points are listed as the reasons why the experimental results cannot be explained in this way. First, the constitutive equation used in this analysis might be inadequate. However, the same results may be obtained for any other constitutive equation under the same assumption, because the elastic terms in constitutive equations usually involve λ of very small magnitude under the condition of the present experiment. Second, questions about the assumption of quasi-steady flow might be asked. When two surfaces begin to move from the stationary state to the normal squeezing flow, the problem has a discontinuous flow history. The same kind of flow history must occur in this experiment, so that unsteady viscoelastic properties such as stress overshoot

may be considered. Phan-Thien et al. reported the numerical analysis of the squeezing flow for a viscoelastic fluid modeled by a constitutive law that allows an overshoot mechanism. They concluded that, essentially, the contribution of the elasticity is due to the stress overshoot which appears at the beginning of the motion, and the steady-state behavior is not very relevant.⁽¹²⁾ Finally, although the flow field was regarded as shear dominant in the above discussion, elongational flow might occur at the beginning of motion because rapid reverse squeezing force was applied to the very thin liquid film.

It is necessary to consider the effect of the above points in order to solve the reverse squeezing flow problems of this kind.

4. Conclusions

Experiments have been performed on the reverse squeezing flow of dilute polymer solutions utilizing a flat plate and a spherical surface. The center clearance between two surfaces has been examined where constant forces were applied. The following points were clarified:

(1) Dilute polymer solutions in which the effect of elasticity is not shown under the condition of normal squeezing flows show slower separating rates under light loading than Newtonian fluids. This trend is clear when the separation is small.

(2) The experimental results of dilute polymer solutions can be correlated with the same normalized factors that are used in the Newtonian analysis, but are affected by the concentration of polymer solutions (i.e., the elasticity of solutions).

(3) The results predicted by the theory of inelastic fluids under the assumption of the quasi-steady state agree well with the experimental results. Under the same assumption, however, the smallness of the rate of separation cannot be explained because of the small magnitude of elastic terms in the constitutive equations used.

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