Award Lecture Article

Numerical and Experimental Analyses of Unsteady Viscoelastic Flow in Complex Flow Field

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This paper includes three topics of the unsteady flow analysis of viscoelastic fluids with both numerical and experimental methods, *i.e.* startup flows in channels, three-dimensional flows, and viscous fingering in Hele-Shaw cells. The first topic presents results of the numerical analysis of startup flows using the Leonov model and results of velocity measurements with laser Doppler velocimetry in abrupt contraction flows. The mechanism of transient change in velocity field was discussed in view of the relation to the stress field. In the second topic, the flow of polymer solutions in a rectangular channel with a cavity is considered. Results of flow visualization and the numerical simulation using the Phan-Thien Tanner model are shown. The relation between a three-dimensional flow in the cavity and elastic properties of viscoelastic fluids was found. Finally, in the third part, the unstable growth phenomenon of the interface between two fluids, viscous fingering, is treated. Some examples of anomalous behavior of fingers in surfactant solutions are presented.

Key Words: Viscoelastic fluid / Unsteady flow / Viscous fingering / Numerical simulation / Experiment

1. INTRODUCTION

We often encounter unsteady flows of viscoelastic fluids such as polymer melts and solutions in polymer processing. The flow behavior of polymeric liquids affects both the quality of products and process efficiency. Because transient rheology of viscoelastic fluids is complex, the transient flow of viscoelastic fluids in complex flow fields is a very complicated problem. Consequently, the unsteady viscoelastic flow is an interesting research issue, and it is useful in a practical point of view to understand the mechanism of unsteady viscoelastic flows in complex flow fields.

The author has studied unsteady viscoelastic flows both numerically and experimentally. The present paper includes three topics, *i.e.* the startup flow of viscoelastic fluids, the three-dimensional viscoelastic flow, and viscous fingering in polymer and surfactant solutions. The results of these studies are introduced below in this order.

2. STARTUP FLOW OF POLYMER SOLUTIONS

In polymer processing, we sometimes encounter a startup flow, where a fluid at rest suddenly begins to flow. The measurement of transient responses of fluids in rheometric

startup flows such as a simple shear flow and a uniaxial elongational flow gives us important rheological data. Transient rheology of fluids is utilized for the analysis of the flow behavior in complex flow fields. In practical point of view, the behavior of viscoelastic fluids in complex flow fields relating to flows occurring in polymer processing is important. The velocity and stress fields grow with time and hence the flow behavior of polymeric liquids in a complex flow field is complicated.

Here, we introduce the results of finite element analysis of viscoelastic startup flows¹⁻⁷⁾ using the Leonov model^{8,9)} as a constitutive equation and experimental results of velocity measurements¹⁰⁾ of startup flow of polymer solutions with a laser Doppler velocimetry (LDV).

2.1 Numerical Simulation

The numerical simulation is an effective tool for analyzing the viscoelastic flow in complex flow fields because it provides us with useful information that may be difficult to obtain from experiments, *e.g.* growth of stress field with time. We numerically analyzed two-dimensional flows in contraction, expansion, and contraction-expansion channels with the SUPG (Streamline Upwind Petrov-Galerkin) finite element method.^{11,12)} In the present paper, we show the results

of the flow in 4 to 1 to 4 abrupt contraction-expansion channels, which consist of upstream and downstream wide conduits connected by a narrow one. In this simulation, we considered three channels that are different in the length of narrow channel.

In an abrupt contraction-expansion channel, vortices appear at both upstream and downstream salient corners. In a Newtonian flow at low Reynolds numbers, the size of vortices reaches to a steady state size just after the onset of the flow and is similar in both upstream and downstream regions. On the other hand, in viscoelastic flows, we can see different growth phenomena of vortices. Just after the onset of the flow, the size of the upstream vortex is similar to that of the downstream one. However, the upstream vortex grows with time to reach a steady state, while the downstream vortex decreases with time. The former phenomenon relates to the elongational flow upstream of the contraction¹⁻³⁾ and the change in vortex size with time has a role as the stress relief mechanism.¹³⁾ The latter phenomenon is well known as a Barus effect. We analyzed also the stress field and confirmed the relation between the Barus effect and elastic properties of fluid.

Furthermore, the analysis of stress power elucidates this relation. The stress power w defined by

$$w = \boldsymbol{\sigma} : \mathbf{D} \quad (1)$$

means the work of stress per unit volume and unit time.¹⁴⁾ Here, $\boldsymbol{\sigma}$ is the stress tensor and $\mathbf{D}$ is the rate of deformation tensor.

In the Leonov model, the stress tensor is divided into the viscous term w_v and the elastic term w_e^k defined by

$$w_v = 2\eta_0 s \mathbf{D} : \mathbf{D}, \quad (2)$$

$$w_e^k = \frac{\eta_k}{\lambda_k} \mathbf{C}^k : \mathbf{D}, \quad (3)$$

where η_0 is the zero-shear-rate viscosity, s is a model parameter. η_k and λ_k are viscosity and relaxation time of the k th mode, respectively. $\mathbf{C}^k$ is the Finger tensor of the k th mode. Hence, one can separately analyze viscous effects and elastic effects on the transient change in flow field. It is an advantage of numerical simulations with the Leonov model.

Figure 1 shows the change in the distribution of $w_e \equiv w_e^1$ at the Weissenberg number $We = 3.3$. In the region of $w_e > 0$, the elastic energy is stored into a fluid and contrary in the region of $w_e < 0$ the stored elastic energy is released. As shown in the figure, the fluid stores elastic energy upstream of the contraction and released elastic energy downstream of the expansion. The positive w_e region spreads with time upstream

of the contraction because of the stress growth due to elongational flow near the contraction. On the other hand, just down stream of the expansion, the negative w_e region spreads with time. This result indicates that the flow pattern changes to widen the region for releasing the stored elastic energy. When the length of narrow conduit is short, the Barus effect appears significantly because there is not enough distance for releasing the stored elastic energy in the narrow conduit.

2.2 Velocity Measurement Experiments

In the velocity measurement experiments of startup flow,¹⁰⁾ we considered a 4 to 1 abrupt contraction channel with a rectangular cross section: 16 mm in height and 60 mm in width in the upstream part, and 4 mm in height and 60 mm in width in the downstream part. The test fluids are aqueous solutions of polyacrylamide (PAA, SANFLOC AH-70P supplied by Sanyo Chemical Industries), which has shear-thinning viscosity and strong elasticity. The velocity response was measured with a LDV.

Figure 2 shows the profile of velocity in the flow direction (z) near the entrance to the contraction ($z = 0$) for a 1.0 wt% aqueous solution of PAA at $We = 17$ and $Re = 0.18$, where Re is the Reynolds number. At $t = 0.8$ s, just after the startup of flow, the velocity is largest at the center of channel ($x = y = 0$) and takes a convex profile for each position of z . The center velocity decreases with time and the velocity profile becomes concave. In addition, the growth of vortex region, where the velocity is small, is observed lower right in Fig. 2a.

The mechanism of transition of the velocity profile is as follows: The elongational viscosity increases near the center just upstream of the entrance to the contraction because of the stretch-thickening property in viscosity. As a result, the

Fig. 1. Distribution of stress power w_e at $We = 3.3$.

velocity decreases in this region and hence the velocity profile changes to a concave shape.

The results of the velocity measurement indicate that highly three-dimensional behavior appears in transient flows of viscoelastic fluids. This fact motivated us to numerically simulate three-dimensional viscoelastic flows.

3. THREE-DIMENSIONAL FLOW OF POLYMER FLUIDS IN CHANNELS

Three-dimensional flow property appears remarkably in viscoelastic flows compared to that for Newtonian flows. In general, it is very difficult to realize an unsteady flow that is approximated as a two-dimensional flow as indicated in 2.2. Consequently, the three-dimensional flow analysis is necessary to elucidate the mechanism of transient flow of viscoelastic fluids through complex geometries.

We have numerically analyzed viscoelastic flows through three-dimensional channels such as a rectangular abrupt contraction channel¹⁵⁾ and a rectangular channel with a cavity.¹⁶⁾ We employed the Giesekus model^{17,18)} and the Phan-Thien Tanner (PTT) model^{19,20)} as a constitutive equation. In the present paper, we will introduce the results of viscoelastic flows through a rectangular channel with a cavity. In this study, experiments were also carried out, and the experimental results are indicated below.

Fig. 2. Change in velocity profile with time in a startup flow of a PAA solution upstream of the entrance to contraction.

Figure 3 shows the schematic diagram of a three-dimensional rectangular channel with a cavity and the coordinate system used in both numerical simulations and experiments. Here the flow direction is z , and $H = 5$ mm in the experiments.

Firstly, we show the results of numerical simulation. In the numerical simulation, the finite volume method coupled with the EVSS (Elastic-Viscous Stress-Splitting) scheme^{12,21)} was adopted to solve governing equations of flow and the constitutive equation.

Figure 4 shows path lines at $We = 2$, $Re = 0.02$, and $\varepsilon = 0.001$, where ε is a parameter of the PTT model. Figure 5 shows the projection of path lines onto the $y-z$ and the $x-y$ planes. The swell of main flow near the cavity is observed. In the cavity, highly three-dimensional flow behavior is seen. That is, the fluid near the side wall moves to the center plane in a spiral manner. This prediction qualitatively agrees with the

Fig. 3. Schematic diagram of a three-dimensional rectangular channel with a cavity and the coordinate system.

Fig. 4. Path lines at $We = 2$, $Re = 0.02$, $\varepsilon = 0.001$: (a) main flow, (b) inside the cavity.

experimental results, which are shown below. The spiral flow appears remarkably at high Weissenberg numbers and does not occur in the Newtonian flow.

Figure 6 shows the distribution of normal stress τ_{yy} in the cavity. The region where the normal stress looks towards the center of channel is wide for a viscoelastic fluid compared to that for the Newtonian fluid, and the magnitude of the normal stress is larger for viscoelastic fluids than the Newtonian fluid. Results at other values of We indicate that the spiral flow in the cavity relates to elastic properties of fluids.

In the experiments, we visualized the flow with both the particle tracer method and the dye streak method. Figure 7 shows the path lines in the center plane, xz plane of $y = 0$, under two conditions: $We = 2.3$ and $E = 420$, and $We = 5.1$ and $E = 410$, where E is the elastic number defined by $E = We/Re$. In both cases, swell of the main flow is observed just downstream of the entrance to the cavity. In our experiments¹⁶⁾, it was found that the main flow retreated into the cavity more deeply at higher elastic numbers. It is because the elastic effect

is strong relative to that of inertia at high elastic numbers and normal stress effect increases. The flow pattern is not symmetric and the position where the flow retreats most deeply shifts upstream in viscoelastic flows.

Figure 8 shows results of flow visualization with the dye streak method. The figure shows the flow in the cavity: the flow into the cavity near the sidewall moves towards the center plane in a spiral manner. The present numerical simulation predicted this phenomenon and this fact indicates that the present numerical scheme is appropriate for the simulation of three-dimensional viscoelastic flows.

Fig. 5. Projection of path lines at $We = 2$, $Re = 0.02$, $\varepsilon = 0.001$: (a) yz plane (main flow), (b) yz plane (inside the cavity), (c) xy plane.

Fig. 6. Distribution of non-dimensional normal stress $\tau_{yy}/(\eta_0 W/H)$ in the $y-z$ plane inside the cavity ($-5 \leq y/H \leq 5$, $0 \leq z/H \leq 1$) at $We = 2$, $x/H = 2$.

Fig. 7. Path lines in the xz plane of $y = 0$: (a) $We = 2.3$, $E = 420$, (b) $We = 5.1$, $E = 410$. The flow is from down to up.

Fig. 8. Streak lines in the cavity at $We = 6.4$, $E = 37$. The main flow is from down to up and time passes from top to bottom.

4. GROWTH PHENOMENON OF VISCOUS FINGERS IN POLYMER SOLUTIONS AND IN SURFACTANT SOLUTIONS

A Hele-Shaw cell consists of two parallel plates separated by a narrow gap. When a less viscous fluid is injected into a more viscous fluid in a Hele-Shaw cell, the interface between the two fluids forms a finger-like pattern, which is called a viscous finger. Phenomena relating to the viscous finger can be seen in polymer processing such as the gas-assisted injection molding.

In general, the interface grows with repeating three growth patterns, *i.e.* spreading, splitting, and shielding.²²⁻²⁴⁾ When a fluid with the shear-thinning viscosity such as polymer solutions is used as a high viscous fluid, the effect of shielding appears strongly compared to the counterpart in Newtonian fluids.^{25,26)} For shear-thinning fluids, the apparent viscosity is smaller near a finger tip that grows faster than other fingers because the shear rate is larger than the others. Hence the finger tip grows more than other fingers and the shielding effect appears more strongly.

Figure 9 shows the relation between the tip velocity v_{tip} and the pressure gradient at the finger tip $\Delta p/\ell$.²⁷⁾ The tip velocity non-linearly depends on the pressure gradient and the growth rate in v_{tip} increases with increasing pressure gradient because of the shear-thinning viscosity. In addition, we found that fingering patterns have a fractal structure when fingers are thin and branch enough.²⁵⁾

We also numerically analyzed the viscous fingering.²⁸⁾

Fig. 9. Relation between the tip velocity v_{tip} and the pressure gradient at the finger tip $\Delta p/\ell$.

Figure 10 shows an example of the numerical simulation of viscous fingering in a polymer solution in a Hele-Shaw cell. In this simulation, we used the VOF (Volume-Of-Fluid) method^{29,30)} to represent the interface between two fluids and expressed the non-Newtonian viscosity using the Carreau model. Governing equations were numerically solved with a finite element method.

The numerical simulation can describe the effect of shear-thinning viscosity and predict that the shear-thinning viscosity emphasized the shielding effect. However, the improvement in both expression of interface and resolution in space will be necessary for simulating complicated fingering structures, which are sometimes encountered in experiments.

In recent study of viscous fingering in surfactant solutions,³¹⁾ we found an anomalous motion of finger, which has not been observed in polymer solutions. The test fluids are aqueous solutions of cetyltrimethylammonium bromide (CTAB) with excess of NaSal as a counter ion. The ratio of mol concentration of CTAB (C_d) to that of NaSal (C_s) is kept to 1 to 7.7. Two test fluids that are different in C_d were employed, *i.e.* $C_d = 0.04$ and 0.03 mol/l. For both fluids, sudden protrusion of a finger tip was observed as shown in Fig. 11. Fingers firstly grow with repeating spreading, splitting, and shielding. In general, a finger tip spreads to make the curvature of interface small for stabilizing the interface. However, the finger tip is cuspidate and the spreading does not occur. After a while, the finger tip suddenly protrudes as shown in Fig. 11. In addition, in Figs. 11(b) and 11(c), surface instabilities are also observed on both sides of the finger. Lowering of surface tension on the lateral surfaces of finger is a possible cause of this instability. Because the finger moves parallel to the cell wall and the surrounding surfactant solution flows along the lateral surface, surfactants accumulate on the lateral surfaces and hence the surface tension tends to be small.

Analyzing the relation between the average pressure gradient at the finger tip $\Delta p/\ell$ and the wall shear rate $\dot{\gamma}_{wall}$, we found the sudden protrusion occurred at a critical shear rate.

Fig. 10. An example of numerical simulation of viscous fingering in a rectangular Hele-Shaw cell. The finger grows from left to right. A contour indicates an interface at certain time.

Figure 12 shows the results for CTAB/NaSal in water of $C_s = 0.04$ mol/l, where H is the half cell height.

The critical shear rate is similar to that at a bending point observed in measurements of flow curve using a capillary rheometer.^{32,33)} These results suggest that the sudden protrusion relates to changes in micellar structure and rheological properties due to the flow.

5. SUMMARY

In the present paper, we introduced results of our previous studies of unsteady viscoelastic flows. The transient flow behavior of complex fluids such as viscoelastic fluids, suspensions, liquid crystalline polymers, and emulsions is a

complicated and interesting issue and still has many unresolved problems. We think that combination of numerical and experimental analyses is effective to elucidate the mechanism of various flow phenomena. Furthermore, the analysis of change in fluid inner structures due to a flow is useful because characteristic rheological properties and flow behavior of complex fluids originate from the fluid inner structure. For example, in numerical studies, coupling of micro and macro simulations is one of recent topics in the flow analysis of complex fluids.

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Fig. 11. The growth of viscous finger in a CTAB / NaSal aqueous solution. The images show a part of cell. The finger grows from left to right.

Fig. 12. Wall shear rate $\dot{\gamma}_{wall}$ versus pressure gradient $\Delta p / l$.

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