Numerical Analysis of Viscoelastic Welding Flow
Part 2: Effect of Temperature on Molecular Orientation

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Abstract

In the polymer processing operations of extrusion and blow molding, weld-lines often occur on the product, especially on the parison made by extruding polymer melts. This is because the polymer molecules near the weld-line highly orient owing to the elongational flow and the molecular orientation does not relax. In the present paper, the non-isothermal viscoelastic welding flow in the channel with a spider that supports a mandrel was numerically calculated for analyzing the molecular orientation in the weld-line region. The single-mode Giesekus model was used as a constitutive equation. The effect of the temperature on the velocity, the stress and the molecular orientation in the stress relaxation process at the weld-line was analyzed. The calculations were carried out for the channel wall temperatures T=190, 195, 200, and 205 °C at the inlet temperature T=190 °C. The numerical results showed that the overshoot of the velocity along the centerline downstream of the spider was large when the channel wall temperature was high. For a fluid with remarkable shear-thinning property, the spider with a large rear-end-angle suppressed the overshoot in the case of T=205 °C. When the wall temperature was high, the distance necessary for relaxation of molecular orientation were short, thus little anisotropy remained in the weld region after solidification.

Keywords: Weld-line; Nonisothermal viscoelastic flow; Molecular orientation; Numerical simulation; Giesekus model

1. Introduction

In the polymer processing operations such as the extrusion and the blow molding, weld-lines often occur on the place where separated flows of polymer melts merge. The weld-line remains even on the products and causes the degradation of the mechanical strength of products and the defectiveness in their appearance. When the weld interface is formed, the polymer molecular orientation in the diffusion process causes the degradation of the mechanical and optical properties near the weld-line. Polymer molecules are highly oriented in the direction of the flow near the weld-line region. The mechanical and optical properties degrade in that region if the orientation does not return to a random condition.

The weld-line also occurs in the injection molding. Many studies[1-5] treated the weld-line in the injection molding and the relation between mechanical properties of the weld-line and the processing condition was discussed in the studies. On the other hand, not so many studies treated the weld-line in the extrusion process.

In Part 1[6] of the present study, we numerically analyzed the viscoelastic welding flow around a spider that supported a mandrel under the isothermal steady creeping flow condition. The single mode Giesekus model[7] was applied as a constitutive equation. In the study, we investigated the effect of the spider shape, the rheological property of fluids, and the Weissenberg number on the elongation of molecules. As a result, we found that the elongation rate and stress were decreased near the rear-end of the spider by a simple modification compared to that proposed by Huang et al.[8]. A spider shape with a large spider-rear-end-angle led to the decrease in the elongation rate in the weld region, where the elongational flow is dominant, hence the elongation of molecules was restricted. Moreover, it was indicated that the overshoot of the velocity occurred in the weld region for shear-thinning fluids at high Weissenberg number. The molecular orientation at the channel exit did not vary with the spider shape at high Weissenberg number and did not return to a random condition within the computational region. Especially for highly stretch-thickening fluids, this tendency was strong.

In general, the temperature is one of the important conditions in the polymer processing. The viscosity and the
relaxation time depend on the temperature, hence it also affects both the flow and the molecular orientation of polymer melts. Consequently, we considered the isothermal viscoelastic welding flow to investigate the effect of temperature on both the elongational flow and the molecular orientation in the weld region. In the present study, we numerically calculated the non-isothermal viscoelastic welding flow in channels with a spider used in Part 1. In the previous study, we found the effectiveness of the suppression of molecular elongation due to the modification of the spider-rear-end-angle from $45^\circ$ to $60^\circ$. In Part 2, we investigated the effect of temperature in the channels with a spider whose spider-rear-end-angle was $45^\circ$ or $60^\circ$ under various conditions of the rheological property of viscoelastic fluid and the Weissenberg number.

2. Governing equations

We considered the steady non-isothermal flow of incompressible viscoelastic fluids. When the effects of inertia and gravity are ignored, the equations of continuity, motion, and energy are given by

\[
\nabla \cdot \mathbf{v} = 0 , \\
-\nabla p + \nabla \cdot \tau = 0 , \\
\rho C_p \nabla \cdot \mathbf{v} - \nabla^2 \mathbf{T} + \tau \cdot \nabla \mathbf{v} ,
\]

where \( \mathbf{v} \) is the velocity vector, \( p \) the isotropic pressure, and \( \tau \) the extra stress tensor, \( \rho \) the density, \( C_p \) the specific heat, \( T \) the absolute temperature, and \( k \) the thermal conductivity.

We applied the single-mode Giesekus model as a constitutive equation: The equations are given by

\[
\tau = 2\eta_s D + \tau_p , \\
\tau_p + \lambda \frac{\tau_p}{\eta_p} + \alpha \frac{\lambda}{\eta_p} \tau_p^2 = 2\eta_p D ,
\]

where \( D \) is the rate-of-deformation tensor, \( \eta_s \) and \( \eta_p \) are the solvent viscosity and the polymer viscosity, respectively, \( \alpha \) is the mobility factor and \( \lambda \) is the relaxation time, \( \tau_p \) is the polymeric contribution to the extra-stress tensor, \( \tau_p \) denotes the upper-convected derivative of \( \tau_p \) defined by

\[
\tau_p = \frac{\partial \tau_p}{\partial t} + \mathbf{v} \cdot \nabla \tau_p - (\nabla \mathbf{v}) \cdot \tau_p - \tau_p \cdot (\nabla \mathbf{v}) .
\]

We assumed the relaxation time and the viscosity depended on the temperature according to the following shift factor [9]:

\[
\lambda(T) = a_T \lambda(T_{ref}) , \\
\eta_p(T) = a_T \eta_p(T_{ref}) ,
\]

where \( a_T \) is the shift factor and \( T_{ref} \) is the reference temperature. The temperature dependence of \( a_T \) was estimated by the WLF equation:

\[
\ln a_T = -\frac{c_1(T - T_{ref})}{c_2 + T - T_{ref}} ,
\]

where \( c_1 \) and \( c_2 \) are constant factors.

Similarly to the analysis in Part 1, the molecular orientation tensor was calculated using the following equation:

\[
(1 + 2b)\tau_p = -nH \langle \mathbf{Q} \mathbf{Q} \rangle + nkT\mathbf{\delta} ,
\]

where \( \mathbf{Q} \) is a connector vector which connects two beads of the dumbbell. A material parameter \( b \) is equal to \( \alpha \) of the Giesekus model. This relation was given by Bird et al. [10, 11] in their process of deriving the Giesekus model from the Hookean dumbbell model. \( H \) is the Hookean spring constant, \( n \) the number of beads per unit volume, \( k \) the Boltzmann constant, and \( T \) the absolute temperature. The operator \( \langle \cdot \rangle \) denotes the phase-space average.

3. Numerical methods

We used the Galerkin finite element method for the numerical simulation. For improving both the accuracy of solution and the convergence stability, we employed non-consistent streamline upwind scheme proposed by Carew et al. [12]. An upwinding weight function was applied only to the convective term in the constitutive equation [13]. The mixed method was applied for solving the finite element equations. This method simultaneously calculates the pressure, the velocity, and the extra-stress.

Figure 1 shows the flow diagram of numerical simulation. Firstly, we calculated an isothermal Newtonian flow, i.e. at \( \text{We}=0 \). Next, we calculated the non-isothermal viscoelastic flow at higher \( \text{We} \) using the result of the Newtonian flow as the initial condition. The calculation requires the computation of the temperature field and the iterative computation of the velocity and stress fields. After obtained the converged solution, we continued the calculation at higher \( \text{We} \) using the result obtained as the initial condition until the solution at a target Weissenberg number \( \text{We}^* \) was obtained. The Newton-Raphson method was used for the iteration and the incremental method for the
increment of \( \text{We} \). In the present study, the Weissenberg number was evaluated by \( \text{We} = \frac{\lambda U}{L} \), where \( U \) and \( L \) are the mean velocity and half channel width at \( x = -6 \) mm, respectively and \( \lambda \) is the relaxation time at \( T = T_{\text{ref}} = 463.15 \) K (190 °C).

4. Numerical conditions

The parameters of the Giesekus model used in the simulation at the reference temperature \( T_{\text{ref}} = 463.15 \) K (190 °C) are as follows: \( \alpha = 0.1 \) and 0.45, \( \lambda = 0.43 \) s, \( \eta_p = 140 \) Pa·s, and \( \eta_r = 7360 \) Pa·s. They are the same as the parameters used in the analysis of the isothermal flows in Part 1. Figure 2 shows the steady shear viscosity \( \eta \) and the first normal stress difference \( N_1 \) of the Giesekus model for these parameters and the experimental data at 190 and 205 °C. The steady uniaxial elongational viscosity \( \eta_E \) is also shown in Fig. 3. The shear-thinning of viscosity is strong when \( \alpha \) is large and the stretch-thickening of \( \eta_E \) appears more strongly for smaller \( \alpha \).

As the parameters of the WLF equation that decides the shift factor of the relaxation time, we used \( c_1 = 6.024 \) and
$c_2=508.1$ K. These values were determined using the measurement of dynamic viscoelasticity of LDPE with a cone-plate type rheometer at three temperatures. When these values are applied, the relaxation time becomes short and the viscosity low with the increasing temperature according to Eqs. (7)-(9) because $\alpha_r < 1$ for $T > T_{ref}$. In addition, the following values were applied: $\rho=762$ kg/m$^3$, $C_p=2500$ J/(kg·K), and $\kappa=0.2$ W/(m·K).\cite{14, 15}

The schematic diagram of a channel with a spider is illustrated in Fig.4. In this figure, the boundary conditions and the coordinate system are also indicated, where $u$ and $v$ are the velocities in the $x$ and $y$ directions, respectively and $\tau_{xy}$ is the $xy$ component of $\tau$. The origin is placed at the spider rear end.

The boundary conditions of temperature are as follows: The inlet temperature $T_i$ is 190 °C, and four values of the wall temperature $T_w$ were considered, i.e. $T_w=190, 195, 200,$ and 205 °C.

The numerical simulation was carried out for two spider-rear-end-angles $\theta_2$, i.e. $\theta_2=45^\circ$ ($R_2=5.81$ mm) and 60° ($R_2=3.098$ mm). The spider-front-end-angle $\theta_1$ was fixed to 45° ($R_1=5.81$ mm). Three values of the Weissenberg number $\text{We}$ were used, i.e. $\text{We}=0.48, 0.96,$ and 2.4. Figure 5 shows the finite element mesh in the computational domain, which consists of 1625 nodes and 744 elements. The mesh is the same as that used in the previous paper.

5. Results and discussion

5.1 Temperature field

Figure 6 shows the temperature distribution at $\theta_2=45^\circ$ and $\alpha=0.45$ for three Weissenberg numbers. High temperature regions are observed on the spider wall, on the centerline downstream of the spider rear end, and near the channel wall. The width of high temperature region decreases with increasing $\text{We}$ because the velocity is larger and the diffusion of temperature due to heat conduction is suppressed more greatly at higher $\text{We}$. It is impossible to raise the temperature in the whole channel because the thermal conductivity of polymer melts is small. However, the temperature in the weld region on the centerline increases because of high temperature of the spider wall. The temperature distribution under other conditions of $T_w$ is qualitatively the same as that in Fig.6.

5.2 Velocity field

In Part 1, we indicated that the degree of molecular
orientation in the merging region near the spider rear end became large as the velocity gradient in the flow direction increased in the region\(^6\). Figure 7 shows the distribution of the velocity gradient \(\partial u/\partial y\) around the spider of \(\theta_2=45^\circ\) for the fluid of \(\alpha=0.45\) at \(We=2.4\). Near the wall of the intermediate part of the spider, the area of the large \(\partial u/\partial y\) is wider at \(T_w=205\degree C\) than at \(T_w=190\degree C\). Figure 8 shows the distribution of the velocity gradient in the flow direction (x) \(\partial u/\partial x\) around the spiders of \(\theta_2=45^\circ\) and \(60^\circ\) at \(\alpha=0.45\) and \(We=2.4\). The distributions of \(\partial u/\partial x\) at \(T_w=190\degree C\) and \(205\degree C\) are qualitatively almost the same under the same condition of \(\theta_2\). There are some differences qualitatively. Around the first half of the spider, the region where \(\partial u/\partial x\) is negative and its absolute value is large is wider at \(T_w=205\degree C\) than at \(T_w=190\degree C\). At both the temperatures, the region of large \(\partial u/\partial x\) near the second half of the spider is narrow for \(\theta_2=60^\circ\) in comparison with the region for \(\theta_2=45^\circ\). Downstream of the spider rear end, the elongation flow is dominant and large \(\partial u/\partial x\) is large. The large \(\partial u/\partial x\) regions for each \(\theta_2\) become wider when the temperature rises from \(190\degree C\) to \(205\degree C\). The region for \(\theta_2=60^\circ\) is narrow and lies downstream in comparison with that for \(\theta_2=45^\circ\). The viscosity around the spider wall decreases because of the rise in the wall temperature and the velocity gradient \(\partial u/\partial y\) increases. The area of the large \(\partial u/\partial y\) region is smaller at \(\theta_2=60^\circ\) than at \(\theta_2=45^\circ\).

Figure 9 shows the velocity in the flow direction \(u\) along the centerline downstream of the spider rear end. In all the cases, the increase in \(u\) becomes great with increasing \(T_w\). This is because the viscosity decreases around the spider and the neighborhood of the spider rear end owing to the temperature rise shown in Fig.6. Especially at high Weissenberg number, there is remarkable difference among the results at four \(T_w\). The overshoot of \(u\) is observed at \(\alpha=0.45\) and \(We=2.4\), which is also confirmed in the isothermal analysis. The overshoot at \(T_w=205\degree C\) is the greatest under every condition of \(\theta_2\). The overshoot for \(\theta_2=60^\circ\) is smaller than that for \(\theta_2=45^\circ\). In the case of the fluid of \(\alpha=0.1\), the overshoot is observed at \(\theta_2=45^\circ\) and \(60^\circ\) when
$T_w$ is higher than 195 °C. This phenomenon is not observed in the isothermal flow. At $\alpha=0.1$, there is little difference among the results of each $\theta_2$ differently from the results at $\alpha=0.45$.

Figure 10 shows the velocity gradient $\frac{\partial u}{\partial x}$ on the centerline downstream of the spider rear end. On the centerline, $\frac{\partial u}{\partial x}$ corresponds to the elongation rate. The difference among the maximum values of the elongation rate at each $T_w$ is large at high Weissenberg number in all the cases. This phenomenon appears remarkably for the fluid of $\alpha=0.45$ that has strong shear-thinning viscosity and weak stretch-thickening viscosity. The elongation rate increases consistently with $T_w$. However, at $\alpha=0.45$ and $We=2.4$, the maximum value for $\theta_2=60^\circ$ and $T_w=205$ °C is smaller than the value for $\theta_2=45^\circ$ and $T_w=190$ °C. In addition, for the fluid of $\alpha=0.45$ that has stronger shear-thinning property, the maximum elongation rate at $We=2.4$ for $\theta_2=60^\circ$ and $T_w=205$ °C is almost the same as that for $\theta_2=45^\circ$ and $T_w=195$ °C. These results indicate that one can restrict the growth of the elongation rate using the spider of $\theta_2=60^\circ$ as reported in Part 1 even though the wall temperature increases. Moreover, the maximum value is almost the same as or lower than that for $\theta_2=45^\circ$ at 10 -15 °C lower temperature.

5.3 Stress field

Figure 11 shows the distribution of the normal stress difference $\tau_{xx}-\tau_{yy}$ around the spider at $We=2.4$. The distribution patterns at $T_w=190$ and 205 °C are almost the same for the same $\theta_2$. For both $\theta_2=45^\circ$ and 60°, $\tau_{xx}-\tau_{yy}$ is large around the downstream of the spider rear end where the elongational flow is dominant. The large $\tau_{xx}-\tau_{yy}$ region
Fig. 10 Velocity gradient $\partial u / \partial x$ along the centerline downstream of the spider rear end.

Fig. 11 Distribution of the normal stress difference $\tau_x - \tau_y$ around the spider at $\alpha=0.45$ and $We=2.4$. 

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spreads towards downstream when $T_w$ rises from 190 °C to 205 °C. The region for $\theta_2=60°$ places downstream in comparison with that for $\theta_2=45°$. This result agrees with the result in Fig.8 that the large $\partial u/\partial x$ region spreads towards downstream with the temperature rise.

The distribution of $\tau_{xx}-\tau_{yy}$ along the centerline downstream of the spider rear end is shown in Fig. 12. On the centerline, $\tau_{xx}-\tau_{yy}$ corresponds to the elongation stress. At We=0.48 and 0.96, there is small difference in the elongation stress among the results at every $T_w$ in all the cases. On the other hand, at We=2.4, the maximum elongation stress increases with the rise in $T_w$. In addition, for the fluid of $\alpha=0.45$, the position of the maximum stress moves downstream. This is because the maximum value of $\partial u/\partial x$ increases and the position where $\partial u/\partial x$ takes the maximum shifts downstream owing to the temperature rise. At $\alpha=0.1$ and We=2.4, the maximum elongation stress is lower in the case of $\theta_2=60°$ and $T_w=205 °C$ than that of $\theta_2=45°$ and $T_w=190 °C$. For the fluid of $\alpha=0.45$, the maximum value for $\theta_2=45°$ at $T_w=190 °C$ is similar to that for $\theta_2=60°$ at $T_w=205 °C$. Consequently, the modification of the spider shape from $\theta_2=45°$ to $60°$ is useful for the restriction of the stress growth when the wall temperature rises.

5.4 Molecular orientation (dumbbell orientation)

Figure 13 shows the distribution of the root-mean-square of the dumbbell's length ($Q^2/Q_{0}^2$) along the centerline downstream of the spider rear end, where $Q$ is the dumbbell length and $Q_0$ is the initial dumbbell length. At We=2.4, there is little difference in ($Q^2/Q_{0}^2$) at the channel exit among the results at every $T_w$ for each $\theta$. However, the distances for the relaxation of ($Q^2/Q_{0}^2$) at each $T_w$ are
slightly different for the fluid of $\alpha=0.45$.

Figure 14 shows the corresponding distribution of the degree of orientation of the dumbbells. The degree of orientation is 0 for a random orientation and is 1 for a perfect orientation\cite{4}. The distance for the relaxation of the molecular orientation diminishes as $T_w$ increases because the relaxation time decreases. At $We=0.48$ and 0.96, the orientation relaxes before $x$ is about 20 mm. At $\alpha=0.45$ and $We=0.96$, the temperature dependence of the distance for the relaxation is strong for each $\theta_2$ and the effect of the wall temperature appears remarkably. At higher Weissenberg number, i.e. $We=2.4$, the degree of orientation in the region of $x$ is between 0 and 8 mm increases as $T_w$ rises, while at $x$ is larger than about 8 mm, the molecular orientation relaxes faster at higher $T_w$. The effect of the temperature rise higher than 200 °C is weak at $\alpha=0.45$ and $\theta_2=45^\circ$ in comparison with that for $\theta_2=60^\circ$. The reason of this phenomenon is as follows: The elongation rate increases near the spider owning to the temperature rise. However, except for the case of $\theta_2=45^\circ$ and $\alpha=0.45$, the relaxation of dumbbell’s orientation is accelerated downstream of $x=8$ mm. At $\theta_2=45^\circ$ and $\alpha=0.45$, the increase in the elongation rate is larger than that under other conditions hence the relaxation near the second half of the spider is not enough.

If the distance between the spider rear end and the exit, where the stretched molecules relax, is longer than 30 mm, it is possible to accelerate the relaxation of molecular orientation by raising the temperature even at $We=2.4$. On the other hand, if the distance is less than about 8 mm, the distance is not enough for the relaxation because the temperature rise causes strong stretch of molecules due to the increase in the elongation rate. Thus, the fluid is cooled...
to solidify before the orientation relaxes enough and the anisotropy remains in a product. It should be noticed that this phenomenon tends to occur in the flow of highly shear-thinning fluids of large $\alpha$ at high $We$. Hence enough distance for the orientation relaxation is required at high temperature. At $We=2.4$, the orientation relaxes fastest under the condition of $\theta_2=60^\circ$ and $\alpha=0.45$ because the restriction of the growth of the elongation rate by the spider shape modification intensifies the temperature effect on the orientation relaxation in the case of highly shear-thinning fluids.

In Part 1, we indicated the effect of the modification of spider shape on the restriction of the growth of the elongation rate. The molecular orientation, however, did not relax to a random orientation in the analyzing region at $We=2.4$. The increase in the wall temperature accelerates the relaxation even for the fluid of $\alpha=0.1$ that has strong stretch-thickening viscosity. Moreover the orientation returns to a random condition in the case of the fluid of $\alpha=0.45$.

6. Conclusion

In the present study, we numerically calculated the non-isothermal viscoelastic welding flow using the single mode Giesekus model as a constitutive equation. We analyzed the effects of temperature, rheological properties, the spider shape, and the Weissenberg number on the elongation rate and the elongation stress in the weld region, where the elongational flow is dominant. In addition, these effects on the distance for the molecular orientation were investigated.
Because thermal conductivity of polymer melts is small, it is impossible to raise the temperature in whole the channel. However, it is possible to raise the temperature in the weld region on the centerline where the elongational flow is dominant and to accelerate the relaxation of molecular orientation.

For highly shear-thinning fluids, the overshoot of the velocity $u$ on the centerline in the weld region increases consistently with $We$; The overshoot is restricted using the spider of large spider-rear-end-angle. The elongation rate near the spider rear end increases with the temperature rise. Especially at high $We$, the effect of temperature is strong for highly shear-thinning fluids and the growth of the elongation rate and stress are suppressed by the modification of the spider shape.

The relaxation of molecular orientation in the region from the spider rear end and the channel exit is accelerated by the temperature rise, thus the orientation is relaxed enough and the anisotropy decreases. However, in the case of the spider with a small rear-end-angle, the effect of temperature on the relaxation becomes weak at high $We$ for highly shear-thinning fluids if the temperature is too high.

The die temperature is an important process condition in both the blow molding and the extrusion. The results of the present analysis indicate that the mechanical and optical properties of products made by the polymer processing are improved by controlling the temperature of die. For this control, it is necessary to consider the characteristics of polymer melts to accelerate the relaxation of the molecular orientation due to the temperature rise.

References