Multiscale Simulation of the Flows of a Bidisperse Entangled Polymer Melt

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We have investigated the flow properties of monodisperse and bidisperse entangled linear polymer melts in a contraction-expansion channel. Unlike conventional macroscopic approaches, where the transport equations are combined with a phenomenological constitutive equation for the stress originating from the polymer dynamics, we use a multiscale simulation (MSS) method, in which a macroscopic simulation model for solving the momentum balance equation is connected to a molecular-based mesoscopic model that can describe the entangled polymer dynamics. In the MSS method, we combine the smoothed particle hydrodynamics method with the dual slip-link model. In this study, we newly investigate the flow properties of a bidisperse entangled linear polymer melt since the polymer melts used in most industrial processes are not monodisperse. Compared with constitutive equations, the slip-link model employed in our MSS is effective at addressing the rheological properties of bidisperse entangled polymer melts. We especially focus on the flow properties of an entangled polymer melt containing a relatively small number of longer polymer chains. The MSS approach enables the relations between the macroscopic complex flows and microscopic states of bidisperse entangled polymer chains to be investigated. As a result of the MSSs, we observe that the flow rate for the bidisperse melt induced by an externally imposed pressure difference is clearly smaller than that for a monodisperse melt because of the high stress originating from the long chains.

Key Words: Multiscale simulation / Slip-link model / Smoothed particle hydrodynamics / Bidisperse polymer melt

1. INTRODUCTION

Entangled polymer melts are known to have a spatiotemporal hierarchical structure, and computer simulations are powerful tools for addressing the flow problems of such entangled polymer melts.1,2) To date, various molecular and phenomenological models have been developed to describe the dynamics at different scales of interest. However, it is clear that the dynamics at lower (microscopic) levels affect the dynamics at higher (macroscopic) levels, and vice versa. Consequently, to accurately predict macroscopic polymeric flows, lower (microscopic)-level dynamics that affect macroscopic flow properties should be taken into account.

There are several approaches to observe microscopic information and macroscopic flow properties. A common method is to employ coarse-grained molecular dynamics simulation techniques, such as the Kremer-Grest model,3) to perform long-term simulations at large spatial scales. However, although this model can evaluate the detailed microscopic information of entangled polymer chains, its large computational cost does not allow the macroscopic flow properties needed for industrial polymer processes to be acquired. Another approach is to develop molecular-based constitutive equations. Unfortunately, while considerable efforts have been made to develop molecular-based constitutive models, such as the Rolie-Poly model for the rheology of monodisperse entangled linear polymer melts,4) it remains difficult for such models to address the rheology of industrial polymer melts (only recently was the Rolie-Poly model extended to address polydisperse polymer melts5). Moreover, the conventional approaches using phenomenological constitutive equations prohibit the microscopic information of polymer chains from being retrieved.

A promising approach for addressing macroscopic flows of entangled polymer melts is the multiscale simulation (MSS) technique. In the MSS technique, instead of employing constitutive equations, a microscopic or mesoscopic simulation model is used to obtain the polymeric stress. MSSs have two main advantages over conventional constitutive model-based flow predictions. One of the advantages is that MSSs allow the microscopic information of polymer chains to be evaluated; the second is that the application of the MSS technique to complex systems such as polydisperse entangled polymer melts and suspensions is relatively straightforward if an adequate microscopic (or mesoscopic) model is utilized. Based on the pioneering MSS studies for polymeric flows by Murashima and Taniguchi6,7) we extended an MSS technique for entangled polymer melts to address the melt spinning process9) flows in a contraction-expansion channel,10) and nonisothermal flows...
in coaxial cylinders. Recently, Seryo et al. developed a machine learning MSS approach based on the Gaussian process, a Bayesian learning approach (although this technique has yet to be applied to entangled melts). To date, all of the above MSS studies have been limited to monodisperse (entangled) polymer melts.

As mentioned above, the advantage of the MSS approach over constitutive models is that the mesoscopic (slip-link) model employed in the MSS is effective for addressing the rheological properties of entangled melts with a molecular weight distribution. In this study, we examine the flow of a bidisperse entangled linear polymer melt in a contraction-expansion channel utilizing our MSS approach, the details of which are shown below.

2. MODEL

The MSS method used in this study is based on the method originally developed by Murashima and Taniguchi. For a macroscopic flow simulation, we use the smoothed particle hydrodynamics (SPH) method, in which a fluid is discretized into \( N_i \) fluid particles, and the governing equations are solved for each fluid particle. The basic governing equations for fluid particle \( i \) (\( 1 \leq i \leq N_i \)) under isothermal conditions are as follows:

\[
\frac{dx_i}{dt} = v_i, \quad (1)
\]

\[
\frac{dv_i}{dt} = \left( \frac{1}{\rho} \nabla \cdot (\sigma - PI) \right)_{x_i} + F_i, \quad (2)
\]

\[
\frac{d\rho_i}{dt} = - (\rho \nabla \cdot v)_{x_i}, \quad (3)
\]

where \( \rho, \sigma, P \) and \( v \) are the density, stress tensor, pressure and velocity field, respectively, \( x_i, v_i \) and \( \rho_i \) are the position, velocity, and density of fluid particle \( i \), respectively, and \( F_i \) is the external body force density on the fluid particle. The stress tensor at particle \( i \), \( \sigma_i \), contains two contributions and can be written as \( \sigma_i = \sigma_i^{(p)} + \sigma_i^{(d)} \), where \( \sigma_i^{(p)} \) is the polymeric stress and \( \sigma_i^{(d)} \equiv \eta^{(d)} (\nabla v + \nabla v^T) \) is the dissipative stress originating from the friction between segments on a smaller spatial scale than the average distance between two consecutive entanglements on polymer chains. Here, \( \eta^{(d)} \) is the Newtonian viscosity. Here, the notation \( ( \cdots )_{x_i} \) is used to show the physical quantity \( ( \cdots ) \) at fluid particle \( i \) obtained by the SPH interpolation. We also use the notation \( ( \cdots )_{x_i} \) to indicate that the SPH interpolation of the physical quantity \( ( \cdots ) \) is performed at the position of fluid particle \( i \).

The above three equations are numerically solved under incompressible conditions, i.e., \( (\nabla \cdot v)_{x_i} = 0 \). To perform macroscopic simulations under incompressible conditions, we employ the equation of state that relates density to pressure. That is, the so-called weakly compressible SPH method is utilized. This is a typical approach for flows with a low Reynolds number, including flows of viscoelastic liquids.

There are two possible ways to calculate the density in the SPH method. One method is to compute the density of fluid particle \( i \) by the weighted sum over the surrounding particles as \( \rho_i = \sum_j m_j W_{ij} \). Here, \( m_j \) is the mass of the fluid particle \( j \), and \( W_{ij} \) is the kernel function. The other method is to directly solve the equation of continuity (eq (3)) using the SPH interpolation method. In this study, we employ the latter option. We show in the next section that the density remains almost constant even if we use this method. This approach has a clear advantage over the former method; that is, the former method would poorly evaluate the density near the free surface due to a shortage of surrounding particles. Although we do not address free surface flows in this study, it will be important to employ the flexible method to address free surface flows in future research.

To stabilize the SPH simulations, we employ the following two methods. The first approach is the particle shifting method that is frequently used in modern SPH simulations; we employ the diffusion-based particle shifting method developed by Lind et al. The second approach is the kernel gradient correction method developed by Oger et al. In this study, to avoid incurring a computational cost higher than that of the basic SPH method, we do not employ the modified SPH method used in the original MSS method. It should be noted that the method for stabilizing the SPH simulations has not been fully established. Therefore, a trial-and-error approach must be taken to determine which method to use.

In this study, we consider a contraction-expansion channel, which is one of the test geometries of flows in the polymer industry. Similar to our previous study, the periodic boundary condition is employed in the horizontal direction. Since longer chains of bidisperse entangled melts have very long relaxation times, it is difficult to reach a steady state; therefore, we focus on the start-up behavior of flows in the contraction-expansion channel. For the wall boundaries, we use the no-slip boundary condition following our previous study.

For a microscopic polymer dynamics simulation to obtain the stress \( \sigma_i^{(p)} \) generated by the entangled polymer dynamics, we use the dual slip-link model. In this study, the effect of friction reduction in flows under high strain rates is not considered because the method to incorporate the friction reduction into the slip-link model for bidisperse entangled melts...
is not fully understood. The effect of the friction reduction on rheological properties of bidisperse melts was studied by Takeda et al.\textsuperscript{21} in which the chain stretch for each component was utilized to determine the degree of the reduction in friction. While this method seems to work to some extent, the friction acting on the polymer chain should be determined by the surrounding chains. Therefore, this method does not appear to be fully rationalized and further studies are still desired.

At the macroscopic level, we use the initial distance between adjacent fluid particles $b$ as the unit length, the mass of a fluid particle $m$ as the unit mass, the Rouse time of a chain with a molecular weight $M_e$ as the unit time, and $\eta_0/\tau_e$ as the unit stress with $\eta_0$ being the zero shear viscosity obtained by $\eta_0 = \eta_0^{(p)} + \eta^{(o)}$. Here, $\eta_0^{(p)}$ is the zero shear viscosity computed by the slip-link model. At the microscopic level, we use the equilibrium length between slip links $a$ as the unit length and $\tau_e$ as the unit time. Moreover, the unit stress of the slip-link model $\sigma_e$ is related to the plateau modulus $G_N$ as $\sigma_e = (15/4)G_N$. While the system size and macroscopic flow parameters are the same as those used in our previous study,\textsuperscript{10} we here briefly explain the system size and parameters for readers’ convenience. We consider a $41.14$ contraction-expansion channel whose system size is $(L_x/b, L_y/b) = (80, 56)$ and that of the narrow channel region is $(\ell_x/b, \ell_y/b) = (28, 14)$. Here, the $x$- and $y$-direction are the horizontal and vertical direction, respectively. Since the length of the $z$-direction is assumed to be infinite, a flow in this study can be considered as a two-dimensional flow. Flows are induced by applying the external body force density $F_i/(\eta_0/\tau_e) = (7.5 \times 10^{-4}, 0)$. All of the simulations presented in this article are performed by self-developed codes.

### 3. RESULTS AND DISCUSSION

#### 3.1 Rheological Properties

Before discussing the results of the MSSs, we present the basic rheological properties obtained by the slip-link model. In this study, as shown in Table I, we focus on two different entangled polymer melts: melt (i) is a monodisperse entangled linear polymer melt with $Z_0 = 10$ ($\sim 93$ wt\% or $Z_0 = 37$ ($\sim 7$ wt\%). Here, $Z_0$ is the number of entangled strands per polymer chain. The molecular weight (the number of entanglements) of each component in the bidisperse melt (melt (ii)) is the same as that in the famous experimental work by Nielsen and coworkers,\textsuperscript{22} but the weight fraction is different from that examined in the experiment. We choose the hypothetical bidisperse system because the objective of this study is to show applicability of the MSS method to bidisperse systems. Figure 1 (a) shows the linear viscoelasticity data obtained from equilibrium simulations using the multiple tau correlator:\textsuperscript{23} From Fig. 1 (a), the average relaxation times for the two melts, $\langle \tau^{(i)} \rangle$ and $\langle \tau^{(ii)} \rangle$, can be determined to be $\langle \tau^{(i)} \rangle \approx 4.5 \times 10^2 \tau_e$ for melt (i) and $\langle \tau^{(iii)} \rangle \approx 1.4 \times 10^3 \tau_e$ for melt (ii). These average relaxation times are calculated using the multimode fitting

### Table I Entangled polymer melts considered in this study.

| Parameters                  | Melt (i) | Melt (ii) |
|-----------------------------|---------|----------|
| No. of short chains ($Z_{0,short} = 10$) | 1000    | 980      |
| No. of long chains ($Z_{0,short} = 37$) | 20      | 20       |
| Average relaxation time: $\langle \tau \rangle / \tau_e$ | $4.5 \times 10^2$ | $1.4 \times 10^3$ |
| Zero shear viscosity: $\eta_0^{(p)} / \sigma_e \tau_e$ | 62.6    | 92.0     |

Fig. 1 Rheological properties of monodisperse (open symbols) and bidisperse (closed symbols) entangled polymer melts. (a) Storage (circles) and loss (diamonds) moduli, (b) transient shear viscosities and (c) transient planar extensional viscosities. In (b) and (c), the black solid and red dashed lines correspond to the linear viscosity growth functions for monodisperse and bidisperse melts, respectively. Here, the linear viscosity growth function for shear flows is $\eta^s(t)$, and that for planar extensional flows is $4\eta_e(t)$. In (b) and (c), the dimensionless strain rates $\gamma \tau_e$ or $\dot{e} \tau_e$ are $4.5 \times 10^{-6}$ (squares), $1.5 \times 10^{-3}$ (circles), $4.5 \times 10^{-3}$ (triangles), and $1.5 \times 10^{-2}$ (diamonds).
The ranges of $\dot{\gamma}$ and $\dot{\omega}$ for melt (ii) are larger than those for melt (i). Figure 1 (c) shows the transient planar extensional viscosities $\eta_p(t)$ for strain rates in the range $\gamma \tau_e \leq 1.5 \times 10^{-3}$ for melt (i) and $\gamma \tau_e \leq 4.5 \times 10^{-4}$ for melt (ii). At high strain rates, however, the planar extensional viscosities show nonmonotonic behavior because we do not consider the effect of friction reduction.

### Table II Density values obtained from the MSSs.

| Melt (i)  | Melt (ii) |
|-----------|-----------|
| Maximum density 1.002 | 1.002 |
| Minimum density 0.9969 | 0.9974 |
| Maximum fluctuation 0.31 % | 0.26 % |

In this section, we explain the results of the MSSs. First, we describe the macroscopic properties obtained from the MSSs. We first note that the density fluctuation is sufficiently suppressed. We compute the maximum and minimum density every 200 steps ($= 1 \tau_e$) during the simulations, and we define the maximum and minimum density fluctuations as $100[\rho_{\text{max}}/\rho_{\text{min}} - 1]$, where $\rho_{\text{max}}/\rho_{\text{min}}$ is the maximum/minimum density in the simulation and $\rho_0$ is the initial density ($\rho_0 = 1$). As shown in Table II, the density fluctuations for both melts are less than 1%, satisfying the requirement for incompressible conditions. We can therefore confirm that incompressible conditions can be achieved even if we directly solve the equation of continuity (eq (3)). (We have also checked the simulation results when we employ the formula for the density, $\rho_t = \sum_j m_j W_{ij}$, and have confirmed that the results are qualitatively similar.)

Figure 2 presents the spatial distributions of the $x$-component of the velocity $v_x$ at time $t = 400 \tau_e$ for melt (i) and melt (ii) (color online). As shown in Fig. 2, the velocity in the narrow channel region for melt (ii) is clearly smaller than that for melt (i). Furthermore, the temporal evolution of the $x$-component of the spatially averaged velocity $v_{ave}$ obtained from the MSSs is plotted in Fig. 3. Here, the spatially averaged velocity is defined as $v_{ave} = \sum_{j=1}^{N_f} v^{ij}_x / N_f$. The velocity profiles initially show overshoots for both melts because the polymer chains are fully relaxed before starting the MSSs. Moreover, the overshoot behaviors for both melts are almost identical since their start-up behaviors are nearly indistinguishable, as shown in Figs. 1 (b) and (c). Nevertheless, after the initial overshoot,
the ranges

\[ p, G, \eta \]

As shown in Fig. 1 (b), these two systems qualitatively show the same behavior, although steady-state values of the shear viscosities clearly deviate from the linear viscoelastic behavior. The extensional viscosities show almost the same behavior as the torsion viscosities, satisfying the requirement of the same body force for both melts. The spatially averaged velocity initially checked the simulation results when we employ the formula for strain rates and stresses for both melts below. Furthermore, the temporal evolution of the stress fields for both melts is plotted in Fig. 3. Here, the spatially averaged velocity is obtained from the MSSs along the wall of the narrow channel region. The velocity profiles initially have been fully relaxed before starting the MSSs. Moreover, the over-relaxation of the density fluctuation is sufficient for melt (ii). To clarify the reason for this difference, we show the average velocity of melt (ii) is clearly smaller than that of melt (i). To clarify the reason for this difference, we show the strain rates and stresses for both melts below.

We show snapshots of the (a) xx-component and (b) xy-component of the velocity gradient tensor \( \mathbf{\kappa} \) in melt (ii) at \( t = 400 \tau_c \) in Fig. 4. While the flow along the horizontal centerline is dominated by planar extensional flow, the flow near the wall of the narrow channel region is mainly governed by shear flow. Moreover, these flow types are mixed at the corner of the narrow flow channel. Quantitative explanations for the above statements can be found in the Appendix for a Newtonian fluid. We can confirm that the strain rate observed in the MSSs corresponds to that examined by the bulk slip-link simulations shown in Figs. 1 (b) and (c). We can use the Weissenberg number to characterize the strain rate, and the strain rates are normalized by the Rouse time of either the longer chains (\( \tau_R^{(\text{long})}/\tau_c = (Z_{0,\text{long}})^2 = 1369 \)) or the shorter chains (\( \tau_R^{(\text{short})}/\tau_c = (Z_{0,\text{short}})^2 = 100 \)). In what follows, we use the Weissenberg number as a term to describe the strain rate normalized by the Rouse time. Therefore, if the Weissenberg number is larger than unity, the polymer chains are expected to be stretched. Figure 5 shows (a) the extensional deformation-based Weissenberg numbers along the horizontal centerline (\( y/b = 0 \)) and (b) the shear deformation-based Weissenberg numbers along the wall of the narrow channel region (\( y/b = \pm 6.5 \)). Here, the origin of the macroscopic coordinates is the center of the system, and the walls of the narrow channel region are at \( y/b = \pm 7 \). Black diamonds and red circles signify the strain rates normalized by the Rouse time for longer chains and shorter chains, respectively. Figure 5 (a) indicates that the longer polymer chains along the line \( y/b = 0 \) are stretched near the entrance and exit of the narrow channel region, whereas the shorter chains along the same line remain unstretched (\( \kappa_{xx} \tau_R^{(\text{short})} < 1 \)). On the other hand, as shown in Fig. 5 (b), along the line \( y/b = -6.5 \) and inside the narrow channel region, the shorter chains are expected to be slightly stretched, while the longer polymer chains are expected to be strongly stretched.

As shown in eq (2), the temporal evolution of the velocity is determined by the gradients of the stress and pressure and the applied external body force. Since the applied external body force for the monodisperse melt (melt (i)) is the same as that for the bidisperse melt (melt (ii)), the difference in the average velocity is attributed mainly to the difference in stress. Figure 6 (a) plots the xx-component of the stress tensor along the horizontal centerline (\( y/b = 0 \)), while Figs. 6 (b-I) and (b-II) plot the xx- and xy-components of the stress tensor, respectively, along the wall of the narrow channel region (\( y/b = -6.5 \)). The black circles and red circles are for melt (i) and melt (ii), respectively. Figure 1 (c) demonstrates that the difference in the planar extensional deformation between the two melts becomes evident at long times and large strain rates. Judging from Fig. 6 (a), while the stress of melt (ii) is slightly larger than that of melt (i), the difference remains small at \( t = 400 \tau_c \). Figure 6 (b-I) reveals a relatively large difference between melts (i) and (ii), which is due to the large shear strain rate observed near the wall of the narrow channel region. We also note a large stress \( \sigma_{xy} \) near the entrance and exit in Fig. 6 (b-II); these large stress regions correspond to the regions where several flow types are mixed, as shown in Fig. 4 (and in Fig. 9). Figure 6 shows that the large stress originating from the longer chains affects the velocity field. Moreover, considering the Weissenberg numbers shown in Fig. 5, these large stresses are attributed to the stretching of longer chains.

Next, we discuss the microscopic information obtained.
Fig. 6 Stress profiles at typical positions along the (a) horizontal centerline and (b) wall of the narrow channel region. (a) The $xx$-component of the stress tensor and the (b-I) $xx$- and (b-II) $xy$-components of the stress tensor. Black circles and red triangles are for melt (i) and melt (ii), respectively. The shaded region corresponds to the narrow channel region.

Fig. 7 Spatial distributions of (a) the average chain length $⟨L⟩$ normalized by the equilibrium length $L_{eq}$ and (b) the average number of entanglements $⟨Z⟩$ normalized by the equilibrium value $Z_{eq}$ for the (I) shorter and (II) longer chains in melt (ii) at $t = 400\tau_e$ (color online).

Table III Orientation parameters obtained from the MSSs.

| Position | $\lambda_{\text{max}}^{\text{short}}$ | $\lambda_{\text{max}}^{\text{long}}$ | $\theta^{\text{short}}$ [deg] | $\theta^{\text{long}}$ [deg] |
|----------|-----------------|-----------------|-----------------|-----------------|
| (a)      | 0.71            | 0.79            | 1.8             | 2.2             |
| (b)      | 0.39            | 0.47            | -20.0           | -0.8            |
| (c)      | 0.65            | 0.73            | -85.6           | -85.5           |
| (d)      | 0.82            | 0.91            | 23.8            | 22.8            |
| (e)      | 0.85            | 0.96            | 12.0            | 6.3             |
| (f)      | 0.76            | 0.91            | -14.1           | -19.2           |

from the MSSs. Figure 7 shows the spatial distributions of (a) the average chain length $⟨L⟩$ normalized by the equilibrium length $L_{eq}$ and (b) the average number of entanglements $⟨Z⟩$ normalized by the equilibrium value $Z_{eq}$ for the (I) shorter and (II) longer chains in melt (ii) at $t = 400\tau_e$ (color online).

shorter chains, chain stretching is evident only near the wall of the narrow channel region where the Weissenberg number for these chains is slightly larger than unity. For the longer chains, chain stretching is observed both along the centerline and near the wall of the narrow channel region. As explained previously, these stretching regions correspond to where the Weissenberg number for the longer chains is larger than unity. As shown in Fig. 7 (b), the number of entanglements clearly decreases, especially in the narrow channel region. Moreover, we can see that fluid particles whose the number of entanglements decreases are advected by the outlet flow. The decrease in the number of entanglements under flows can be considered as the effect of the convective constraint release that has already been well documented in molecular rheology. \(^{24}\)

Finally, we present the orientations of polymer chains in...
Fig. 6. Stress profiles at typical positions along the (a) horizontal centerline channel region. The stress tensor. Black circles and red triangles are for melt (i) and (xx) and (b) wall of the narrow channel region. (a) The \( \langle \cdot \cdot \cdot \rangle_{\text{short/long}} \)\( \equiv (x, y, z) \text{), where} \langle \cdot \cdot \cdot \rangle_{\text{short/long}} \)\text{ is the average over short or long chains. The largest eigenvalue} \( \lambda_{\text{max}} \)\text{ of the normalized conformation tensor} \( \mathbf{W}_{\alpha\beta} = \mathbf{W}_{\alpha\beta}/\text{TrW} \)\text{ indicates the degree of chain orientation. Furthermore,} \theta \left( -90^\circ \leq \theta \leq 90^\circ \right) \text{ is the orientation direction with respect to the x-axis as shown in the upper right panel of Fig. 8. Table III shows the maximum eigenvalue and orientation direction for short and long chains. Positions (a)–(f) shown in Table III correspond to those in Fig. 8. Of course, the trend illustrated in Fig. 8 can also be seen in Table III, offering a more quantitative evaluation. As shown in Fig. 4 (and in Fig. 9), the flow type along the horizontal centerline and outside the narrow flow channel is planar extensional flow. At the inlet ((a) \( x/b \approx -14 \)), both the shorter and the longer chains are oriented \( \left( \lambda_{\text{max}} \approx 0.71 \text{ and } \lambda_{\text{max}} \approx 0.79 \right) \text{ in the horizontal direction} \( \langle \theta_{\text{short}} \rangle \approx \theta_{\text{long}} \approx 2^\circ \) in response to the planar extensional flow with \( \kappa_{xs} > 0 \). At the outlet (b) \( (14, 0) \text{), the shorter chains are almost relaxed} \( \left( \lambda_{\text{max}} \approx 0.39 \text{ and } \theta_{\text{short}} \approx -20^\circ \right) \text{, while the longer chains are still slightly oriented in the horizontal direction} \( \left( \lambda_{\text{max}} \approx 0.47 \text{ and } \theta_{\text{long}} \approx -1^\circ \right) \text{ because the longer chains have a larger relaxation time than the shorter chains as noted in the previous section. Beyond the narrow channel region} \text{(c) (25, 0)), both the shorter and the longer chains are oriented} \left( \lambda_{\text{max}} \approx 0.65 \text{ and } \lambda_{\text{max}} \approx 0.73 \right) \text{ in the vertical direction} \( \langle \theta_{\text{short}} \rangle \approx \theta_{\text{long}} \approx -86^\circ \) \text{ in response to the planar extensional flow with} \kappa_{xx} < 0 \text{ and } \kappa_{yy} > 0 \text{. Next, we focus on the flows along the line} y/b = -6 \text{. While the shear and extensional flows are mixed at both the entrance} \text{(d) (x/b, y/b) = (14, -6)) and the exit (f) (14, -6)}, only the shear flow can be detected inside the narrow channel region ((e) (0, -6)). At (d) \text{(14, -6), the chains are strongly oriented} \left( \lambda_{\text{max}} \approx 0.82 \text{ and } \lambda_{\text{max}} \approx 0.91 \right) \text{ to the upper right direction due to the presence of both shear and extensional flows, whereas at (e) (0, -6), the chains are strongly oriented} \left( \lambda_{\text{max}} \approx 0.85 \text{ and } \lambda_{\text{max}} \approx 0.96 \right) \text{ to the upper right direction with smaller angles than those at position (d) (} \langle \theta_{\text{short}} \rangle \approx 12^\circ \text{ and } \theta_{\text{long}} \approx 6^\circ \rangle \text{ due to the strong shear flow. At (f) (14, -6), the polymer chains are still oriented} \left( \lambda_{\text{max}} \approx 0.76 \text{ and } \lambda_{\text{max}} \approx 0.91 \right) \text{, while the orientation direction changes to the lower right direction due to the outlet flow} (\langle \theta_{\text{short}} \rangle \approx -14^\circ \text{ and } \theta_{\text{long}} \approx -19^\circ \rangle) \text{.}

4. CONCLUDING REMARKS

4.1 Summary

In this study, we investigate the flows of monodisperse and bidisperse entangled linear melts in a contraction-expansion channel using an MSS technique. Both macroscopic and microscopic properties are successfully captured. Macroscopically, we observe that the flow rate obtained by the bidisperse melt is smaller than that obtained by the monodisperse melt; this difference is attributed to the large stress originating from the longer chains. The origin of this large stress is the stretching of the longer chains since the strain rates normalized by the Rouse time for the longer chains are considerably larger than unity in the region where large stresses are detected. In addition to the macroscopic information, we present microscopic information, such as the stretching, entanglements, and orientations of the chains. We hope that this MSS approach will help with processing and molecular design in the future.

4.2 Future Perspective

Further development from both microscopic and macroscopic perspectives is needed to improve the MSS method into a more sophisticated technique. For example, Minegishi et al. reported that a polymer melt containing a small amount of a component with an ultrahigh molecular weight exhibits substantial strain hardening under uniaxial extensional flow.\textsuperscript{25} Such a polymer melt should show different macroscopic flow properties from a polymer melt without a component featuring an ultrahigh molecular weight. These flow properties can be detected by our MSS method. In relation to flows of polydisperse polymer melts, another interesting phenomenon would be the chain segregation. This segregation is considered to be related to several macroscopic phenomena, such as wall slip\textsuperscript{26} and melt fracture.\textsuperscript{27} To analyze these phenomena, a concentration field should be taken into account. While the way to address the concentration change has been well established at the macroscopic level, there is no established method to address this effect at the microscopic level. This problem should be considered in our future study. Besides that, we would like to extend the MSS method to predict free surface flows. With the SPH method, it is easier to implement free
surface flows than with Eulerian grid-based methods. After this extension, we can compare macroscopic flow properties, such as the swelling ratio, obtained by the MSS with those obtained experimentally, as Robertson et al. experimentally examined the extrudate swell of monodisperse and polydisperse polystyrenes. It is also necessary to directly compare the microscopic properties obtained by MSSs with those obtained by scattering experiments. For example, Graham et al. performed a scattering experiment on monodisperse entangled linear polystyrene melts passing through a contraction-expansion channel. While the molecular weight considered in this literature is within a range accessible by the MSS, accurately obtaining the structure factor by MSS remains challenging. Microscopically, it would be interesting to incorporate the effect of friction reduction, which is not considered in this study, into the slip-link model for bidisperse entangled melts. These research subjects will be investigated in the future.

**APPENDIX: FLOW TYPE CLASSIFICATION**

To classify the flow types in complex flow geometries, we can use the flow classification parameter $\beta_3$ reported by Giusteri and Seto in which the parameter $\beta_3$ is defined as

$$\beta_3 = \frac{d_3 \cdot \nabla \times \vec{v}}{2 \varepsilon_{\text{eff}}},$$

where $d_3$ is the eigenvector of the strain rate tensor $\mathbf{D}$ with the smallest absolute eigenvalue and $\varepsilon_{\text{eff}}$ is the effective strain rate defined in the main text. The parameter $\beta_3$ has a clear meaning. That is, planar extensional, simple shear, and rigid rotational flows correspond to $\beta_3 = 0$, $\beta_3 = 1$, and $\beta_3 \gg 1$, respectively. For any other $\beta_3$ values, several flow types are mixed.

Figure 9 shows the values of the flow classification parameter $\beta_3$ obtained by a simulation of a Newtonian fluid along the horizontal centerline ($y = 0$) and along the wall of the narrow channel region ($y/b = -6.5$). To calculate $\beta_3$, a Newtonian fluid is used instead of an entangled polymer melt. Although viscoelastic effects are not considered in a Newtonian fluid, this analysis is useful for better understanding the flow types in a contraction-expansion channel. For the flows inside the narrow channel region, the flow type can be classified as shear flow since $\beta_3$ is almost equal to 1. For the flow along the line $y = 0$ and outside the narrow region, the flow type can be classified as planar extensional flow ($\beta_3 \approx 0$). While the flow type along the line $y = 0$ and inside the narrow channel region is classified as shear flow, the shear strain rate along this line is very small as shown in Fig. 4(b). Therefore, we can safely say that the flow along the horizontal centerline is mainly dominated by planar extensional flow. Moreover, it should be noted that slight increase in $|\beta_3|$ at positions $x/b = \pm 40$ along $y = 0$ can be seen. If the length of the wide channel is sufficiently long, the flow type in the wide channel and far from the narrow channel should be classified as shear flow ($\beta_3 \approx 1$). From Fig. 9, we can see that the system size in this study is not large enough to detect a fully developed shear flow in the wide channel. Moreover, for the line $y/b = -6.5$ and near the entrance and exit of the narrow region, several flow types are mixed ($\beta_3 > 1$). More rigorously, we should use the objective flow classification parameter $\bar{\beta}_3$ instead of $\beta_3$ since the flow classification by $\beta_3$ is not objective. This will be performed in our future study.

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Fig. 9 Flow classification parameter $\beta$.

These research subjects will be investigated in the future.

The effect of friction reduction, which is not considered in this study, has a clear meaning. That is, planar extensional, simple shear, and rigid rotational flows correspond to $\beta_1$, $\beta_2$, and $\beta_3$, respectively. For any other case, we can compare macroscopic flow properties, such as the swelling ratio, obtained by the MSS with those reported by Ogasawara Foundation for the Promotion of Science & Engineering.

APPENDIX: FLOW TYPE CLASSIFICATION

Figure 9 shows the values of the flow classification parameter $\beta$. Diamonds are the MSSs presented in the main text, a Newtonian fluid is used to compute the system size in this study is $\bar{b}_0 = 3$. The shaded region corresponds to the narrow channel region ($y_1 = 0$) and along the wall of the narrow channel region ($|y_2| = |y_3| = 0$). While the flow along the horizontal centerline is mainly dominated by planar extensional flow. Moreover, it should be noted that slight increase in the channel region ($y_1 = 0$). Therefore, we can safely say that the flow along the horizontal centerline is mainly dominated by planar extensional flow ($\beta_1$). For the flow along the line $y_1 = 0$, it should be noted that slight increase in $|y_2|$ and inside the narrow channel region $|y_2| = |y_3| = 0$. Moreover, for the line $y_1 = 0$, it should be noted that slight increase in $|y_2|$, $|y_3| = 0$. Therefore, we can safely say that the flow along the horizontal centerline is mainly dominated by planar extensional flow ($\beta_1$).

Viscoelastic effects are not considered in a Newtonian fluid, and outside the narrow region, the flow type can be classified as planar extensional flow ($\beta_1$) flow since $\beta = 3$. For the flow along the line $y_1 = 0$, it should be noted that slight increase in $|y_2|$, $|y_3| = 0$. Therefore, we can safely say that the flow along the horizontal centerline is mainly dominated by planar extensional flow ($\beta_1$).

From Fig. 9, we can see that the system size in this study is very small as shown in Fig. 4 (b). Therefore, we can safely say that the flow along the horizontal centerline is mainly dominated by planar extensional flow ($\beta_1$).

The entrance and exit of the narrow region, several flow types are mixed. Moreover, for the line $y_1 = 0$, it should be noted that slight increase in $|y_2|$, $|y_3| = 0$. Therefore, we can safely say that the flow along the horizontal centerline is mainly dominated by planar extensional flow ($\beta_1$).

To calculate $\beta$, the shear strain rate along this line is not objective. This will be performed instead of $\beta$. For the flow along the line $y_1 = 0$, it should be noted that slight increase in $|y_2|$, $|y_3| = 0$. Therefore, we can safely say that the flow along the horizontal centerline is mainly dominated by planar extensional flow ($\beta_1$).

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