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Effect of flow kinematics on the extensional viscosity of dilute polymer solutions

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Abstract

We investigate the effects of flow kinematics on the extensional viscosities of dilute polymer solutions by conducting dissipative particle dynamics simulations under uniaxial, planar, and biaxial extensional flows. At high extension rates, dilute polymer solutions exhibit strain hardening under these flows, whereas the quantitative behavior depends on the flow type. To elucidate the physical origin of this flow-kinematics dependence, we relate the extensional viscosity to the polymer conformation using an analytical expression derived from a single-chain model. The resulting relation allows us to separate the contribution of flow-induced polymer conformational changes and the purely kinematic contribution determined by the structure of the velocity gradient tensor. When polymers remain almost unperturbed by extensional flows, extensional viscosity differences are governed primarily by purely kinematic effects. In contrast, as the polymers are stretched, the gyration radius in the extensional direction becomes the dominant factor, and differences in the degree of stretching in this direction lead to corresponding variations in the extensional viscosity.

Introduction

Dilute polymer solutions have attracted considerable attention in fluid mechanics because even small amounts of polymer additives can qualitatively alter flow behaviors. Since Toms's experiments that were conducted in the 1940s [1], flexible polymers have been known to drastically suppress turbulence, thereby contributing to drag reduction in wall-bounded flows [2, 3]. In contrast, under laminar flow conditions, polymers can induce elastic turbulence, enhancing the mixing efficiency in microfluidic devices [4, 5]. To elucidate the physical mechanism underlying these intriguing phenomena, a number of researchers have studied flow modulation by polymers through laboratory experiments and numerical simulations based on constitutive equations, such as the Oldroyd-B model and the finitely

extensible nonlinear elastic model with the Peterlin approximation (FENE-P).

In this context, a fundamental understanding of the rheology of dilute polymer solutions is indispensable because it serves as a basis for interpreting complex flow data and assessing the validity of constitutive equations. In realistic situations (e.g., turbulent flows), polymers undergo various flow types, such as shear and extensional flows, as well as their combinations. Moreover, polymer responses are highly sensitive to the nature of the underlying flow kinematics. For example, polymers exhibit significant stretching under extensional flows [6], which contrasts with their behaviors observed under shear flows [7]. Thus, understanding the rheological properties of dilute polymer solutions in different flow types is crucial. In particular, since the shear viscosity of a dilute polymer solution is typically independent of the shear rate or exhibits only weak shear thinning, extensional rheology has been a major focus.

However, owing to the low viscosities of dilute polymer solutions, systematic investigations under extensional flows with different flow kinematics remain scarce [8, 9] compared with those for polymer melts [10–16]. Recently, extensional rheometry techniques for low-viscosity fluids, such as capillary breakup extensional rheometry [17, 18] and cross-slot extensional rheometry [19], have been

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developed, enabling evaluations of the extensional rheology of dilute polymer solutions. Notably, Haward et al. [20] developed a six-arm cross-slot device optimized for generating uniaxial and biaxial extensional flows. Together with a two-dimensional cross-slot device, they systematically investigated the extensional viscosities of dilute polymer solutions under uniaxial ($\eta_E(\dot{\epsilon})$), planar ($\eta_P(\dot{\epsilon}_P)$), and biaxial extensional flows ($\eta_B(\dot{\epsilon}_B)$) [9]. They reported that at low polymer concentrations, $\eta_E(\dot{\epsilon})$, $\eta_P(\dot{\epsilon}_P)$, and $\eta_B(\dot{\epsilon}_B)$ were consistent with the predictions of the FENE-P model, including the relation $\eta_E(\dot{\epsilon}) = \eta_P(\dot{\epsilon}_P) = 2\eta_B(\dot{\epsilon}_B)$ at high extension rates. While these approaches are promising, further work is needed to establish their quantitative reliability, particularly with respect to the nonidealities in the applied flow fields and the approximations used in viscosity calculations.

Molecular simulations can complement experimental measurements and provide valuable insights into the relation between extensional rheology and polymer dynamics. The early studies mainly used Brownian dynamics (BD) simulations to investigate dilute polymer solutions under uniaxial and planar extensional flows [21–28]. These studies have contributed to understanding the behaviors of dilute polymer solutions, from single-molecule stretching to extensional rheological properties. More recently, the dissipative particle dynamics (DPD) method, where solvent particles are treated in an explicit manner, has been employed to investigate dilute polymer solutions under uniaxial extensional flows [29, 30]. In our previous work [30], we conducted DPD simulations of dilute polymer solutions under uniaxial extensional flow and established a quantitative relation between the extensional viscosity and polymer conformation. However, a systematic comparison of the rheological response under different extensional flows, especially under biaxial extensional flows, remains unexplored for dilute polymer solutions, although the effect of biaxial deformation has been studied for polymer melts [31–33]. This is a serious issue because several previous studies have reported that biaxial extensional flows play a crucial role in polymer stretching within turbulent flows [34, 35].

In the present study, we aim to establish a comprehensive understanding of the effects of flow kinematics on dilute polymer solutions. For this purpose, we conduct DPD simulations of dilute polymer solutions under uniaxial, planar, and biaxial extensional flows and compare the extensional viscosities among these flow types. To explain the flow-kinematics dependence of the rheological responses, we relate the extensional viscosity to the polymer conformation. Specifically, we extend our previous approach for uniaxial extensional flow [30], which is based on an analytical relation for a single-chain model, to planar and biaxial extensional flows. The quantitative relations

obtained here enable us to decouple the contributions of flow-induced polymer conformational changes from the purely kinematic effects determined by the structure of the velocity gradient tensor. Consequently, we reveal how flow kinematics affect the extensional viscosities of dilute polymer solutions in terms of polymer conformation.

Method

We employ the DPD method to study dilute polymer solutions. Following previous studies [30, 36], the polymers are modeled as flexible linear chains, each of which consists of N_p particles. Adjacent particles along the chain are connected by a shifted FENE potential U_B :

$$U_B(r) = -\frac{k_F}{2}(r_{\max} - r_{\text{eq}})^2 \ln \left\{ 1 - \left(\frac{r - r_{\text{eq}}}{r_{\max} - r_{\text{eq}}} \right)^2 \right\}, \quad (1)$$

where k_F is the spring constant, r_{eq} is the equilibrium bond length, and $r_{\max}$ is the maximum bond length. In the DPD method, solvent particles are considered explicitly. These DPD particles, including polymer and solvent particles, interact via soft repulsive, dissipative, and random forces. The dissipative force, which depends on the relative velocity of the interacting particles, is computed using the laboratory velocity. Further details regarding the DPD model can be found in previous papers [37, 38]. In the following, all quantities are nondimensionalized using $k_B T$, m , and r_c , where k_B is the Boltzmann constant, T is the temperature of the system, m is the mass of a DPD particle, and r_c is the cutoff distance.

The parameters of the DPD simulations are set as follows: the total number of DPD particles is $N = 648000$; the number density of the particles is $\rho = 3$; the dissipative force coefficient is $\gamma = 4.5$; the random force coefficient is $\sigma = 3$; the number of particles per chain is $N_p = 50$; the volume fraction of the polymer particles is $\phi = 0.1$; the spring constant is $k_F = 40$; the equilibrium bond length is $r_{\text{eq}} = 0.7$; the maximum bond length is $r_{\max} = 2$; and the repulsive force coefficients between different types of particles are $a_{pp} = 25$, $a_{ps} = 25$, and $a_{ss} = 25$, where p and s denote polymer and solvent particles, respectively. We have confirmed that $\phi = 0.1$ is within the dilute regime $\phi < \phi^* (= 0.14)$, where the overlap volume fraction ϕ^* is computed as $\phi^* = N_p / \{(4/3)\pi\rho R_g^3\}$, with R_g being the polymer gyration radius evaluated at $\phi = 0.025$.

We investigate three types of extensional flows. Following previous studies [30, 39], we employ the SLLD equations [40] and the generalized Kraynik–Reinelt (GKR) boundary conditions [41, 42] to generate homogeneous

uniaxial, planar, and biaxial extensional flows. The SLLOD equations are given by

$$\frac{d\mathbf{r}_i}{dt} = \mathbf{p}_i + (\nabla \mathbf{u})^\top \cdot \mathbf{r}_i \quad (2)$$

$$\frac{d\mathbf{p}_i}{dt} = \mathbf{F}_i - (\nabla \mathbf{u})^\top \cdot \mathbf{p}_i, \quad (3)$$

where $\mathbf{p}_i$ is the peculiar momentum of the i -th particle, $\mathbf{F}_i$ is the force acting on that particle, and $\nabla \mathbf{u}$ is the velocity gradient tensor of the imposed flow field. For uniaxial, planar, and biaxial extensional flows, $\nabla \mathbf{u}$ is expressed as follows:

$$(\nabla \mathbf{u})_E = \begin{pmatrix} \dot{\epsilon} & 0 & 0 \\ 0 & -\dot{\epsilon}/2 & 0 \\ 0 & 0 & -\dot{\epsilon}/2 \end{pmatrix}, \quad (4)$$

$$(\nabla \mathbf{u})_P = \begin{pmatrix} \dot{\epsilon}_P & 0 & 0 \\ 0 & -\dot{\epsilon}_P & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad (5)$$

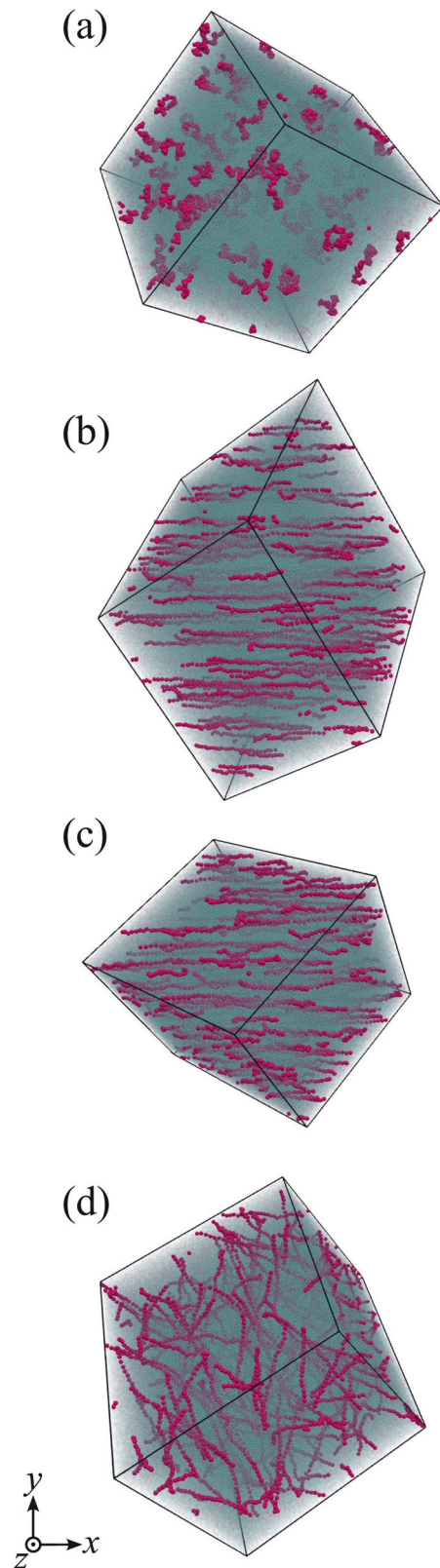
$$(\nabla \mathbf{u})_B = \begin{pmatrix} \dot{\epsilon}_B & 0 & 0 \\ 0 & \dot{\epsilon}_B & 0 \\ 0 & 0 & -2\dot{\epsilon}_B \end{pmatrix}. \quad (6)$$

Here, $\dot{\epsilon}$ is the extension rate, $\dot{\epsilon}_P$ is the planar extension rate, and $\dot{\epsilon}_B$ is the biaxial extension rate. Hereafter, we collectively denote these extension rates by $\dot{\epsilon}_\alpha$. Snapshots of the polymer solution at $t = 0$ (i.e., just before the onset of the imposed flow) and at $\dot{\epsilon}_\alpha t = 15$ in the three extensional flows are shown in Fig. 1. As shown in Fig. 1, the simulation box deforms under the imposed flow. Specifically, the deformation process is governed by the evolution of the lattice basis vectors $\mathbf{e}_i$:

$$\frac{d}{dt} \mathbf{e}_i = (\nabla \mathbf{u})^\top \cdot \mathbf{e}_i. \quad (7)$$

While applying extensional deformation, we systematically remap the deformed simulation box using the GKR method [41, 42] with Semaev's algorithm [43]. To prevent instabilities due to finite-precision arithmetic [44], the total peculiar momentum is periodically reset to zero, as in previous studies [42, 45].

We describe the details of the numerical implementation. The modified velocity Verlet method [46] is used for time integration, where the parameter λ introduced in this scheme and the time step Δt are set to $\lambda = 0.65$ and $\Delta t = 0.04$, respectively. As shown in Fig. 2, the relative temperature error $|k_B T(\dot{\epsilon}_\alpha) - 1|$ increases with $\dot{\epsilon}_\alpha$, where $k_B T(\dot{\epsilon}_\alpha)$ is computed as $k_B T(\dot{\epsilon}_\alpha) = \langle \mathbf{p}^2 \rangle / 3$, with $\langle \cdot \rangle$ denoting the ensemble average. Thus, we determine the range of $\dot{\epsilon}_\alpha$ such that $|k_B T(\dot{\epsilon}_\alpha) - 1| < 0.06$. The initial simulation box is a



cube with dimensions of $60 \times 60 \times 60$. First, equilibrium simulations are conducted from a random initial configuration for 12000 time units, which is sufficiently long

Fig. 1 Snapshots of the polymer solution **a** at $t = 0$ (i.e., just before the onset of the imposed flow) and **b–d** at $\dot{\epsilon}_\alpha t = 15$ in **b** uniaxial, **c** planar, and **d** biaxial extensional flows, with $\dot{\epsilon}_\alpha = 0.0376$ for all the cases. The polymer and solvent particles are shown in red and blue, respectively. For clarity, all the particles are rendered in a semi-transparent manner, except for the selected polymers

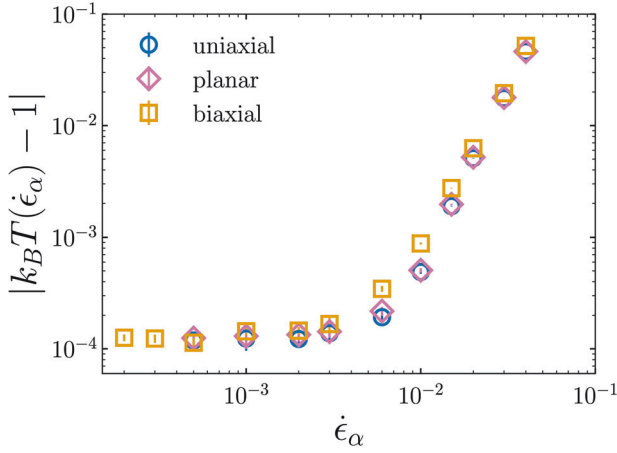


Fig. 2 Relative temperature error $|k_B T(\dot{\epsilon}_\alpha) - 1|$ as a function of the extension rate $\dot{\epsilon}_\alpha$ for uniaxial, planar, and biaxial extensional flows. The error bars denote the standard deviations from ten independent simulations

compared with the longest relaxation time $\tau (= 160)$ of the polymers. After this initial equilibration, we apply extensional flows to the polymer solutions. All DPD simulations are conducted using our in-house code.

Results and discussion

We first investigate the transient response of dilute polymer solutions under start-up extensional flows with different flow kinematics. Uniaxial, planar, and biaxial extensional viscosity growth functions are defined as follows:

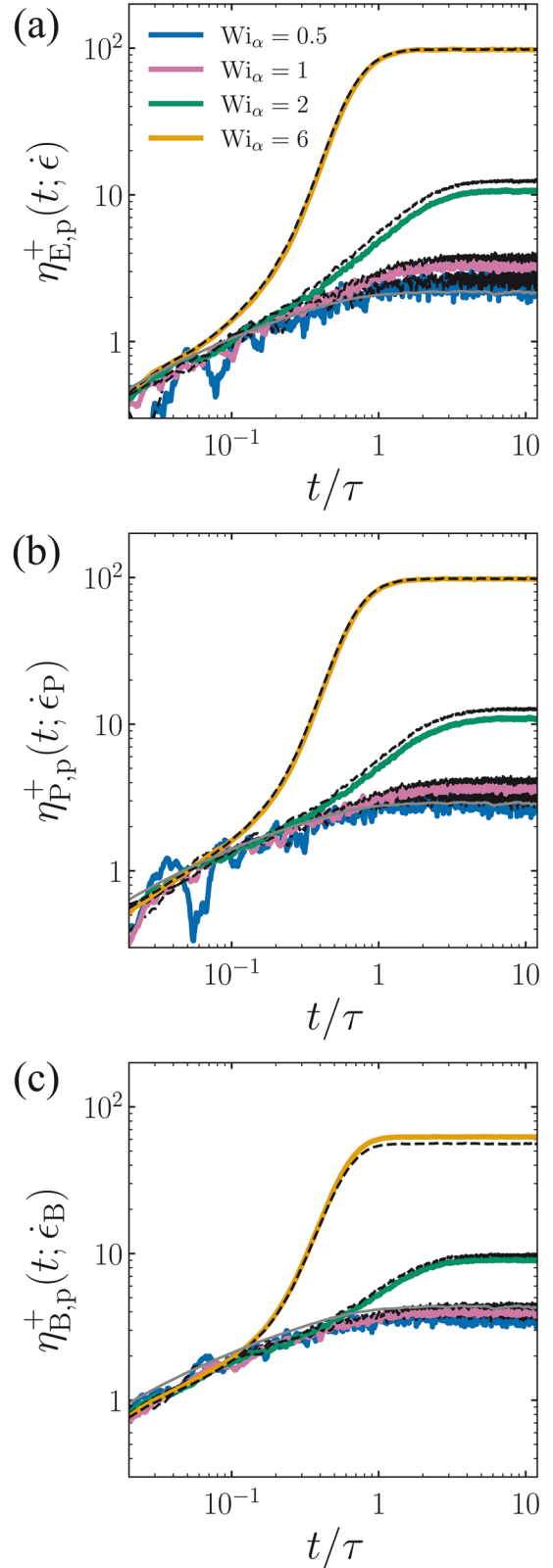
$$\eta_E^+(t; \dot{\epsilon}) = \frac{\sigma_{xx}^+(t; \dot{\epsilon}) - \{\sigma_{yy}^+(t; \dot{\epsilon}) + \sigma_{zz}^+(t; \dot{\epsilon})\}/2}{\dot{\epsilon}}, \quad (8)$$

$$\eta_P^+(t; \dot{\epsilon}_P) = \frac{\sigma_{xx}^+(t; \dot{\epsilon}_P) - \sigma_{yy}^+(t; \dot{\epsilon}_P)}{\dot{\epsilon}_P}, \quad (9)$$

$$\eta_B^+(t; \dot{\epsilon}_B) = \frac{\{\sigma_{xx}^+(t; \dot{\epsilon}_B) + \sigma_{yy}^+(t; \dot{\epsilon}_B)\}/2 - \sigma_{zz}^+(t; \dot{\epsilon}_B)}{\dot{\epsilon}_B}. \quad (10)$$

Here, $\sigma^+(t; \dot{\epsilon}_\alpha)$ is the stress tensor during the start-up extensional flow, which is defined as follows [47, 48]:

$$\sigma^+(t; \dot{\epsilon}_\alpha) = -\frac{1}{V} \left(\sum_{i < j} \mathbf{r}_{ij}(t) \mathbf{F}_{ij}(t) + \sum_i \mathbf{p}_i(t) \mathbf{p}_i(t) \right), \quad (11)$$



where V is the system volume, $\mathbf{r}_{ij}(t)$ is the relative position vector of the i -th particle with respect to the j -th particle, $\mathbf{F}_{ij}(t)$ is the force exerted on the i -th particle by the j -th

Fig. 3 Polymer contribution $\eta_{\alpha,p}^+(t;\dot{\epsilon}_\alpha)$ (thick solid curves) to the extensional viscosity growth function as a function of time t normalized by the longest relaxation time τ of the polymers for **a** uniaxial, **b** planar, and **c** biaxial extensional flows at different Weissenberg numbers Wi_α . The thin gray curves represent the LVE envelopes. The black dashed curves represent the predictions based on the Rouse-type model [Eq. (18)]

particle, and $\mathbf{p}_i(t)$ is the peculiar momentum of the i -th particle. For simplicity, we collectively denote these viscosity growth functions by $\eta_\alpha^+(t;\dot{\epsilon}_\alpha)$. To improve the statistical accuracy of the calculations, we evaluate $\eta_\alpha^+(t;\dot{\epsilon}_\alpha)$ by averaging the results from 20 independent simulations. In addition, we apply a moving average with a window size of $100\Delta t$ to reduce the noise.

We consider the polymer contribution $\eta_{\alpha,p}^+(t;\dot{\epsilon}_\alpha)$ to $\eta_\alpha^+(t;\dot{\epsilon}_\alpha)$ by subtracting the Newtonian solvent contribution, namely, $3\eta_s$, $4\eta_s$, and $6\eta_s$ for uniaxial, planar, and biaxial extensional flows, respectively, where η_s is the solvent viscosity obtained from equilibrium DPD simulations of the solvent using the Green–Kubo formula [40]. Figure 3 shows $\eta_{\alpha,p}^+(t;\dot{\epsilon}_\alpha)$ as a function of the time t normalized by the longest relaxation time τ of the polymers. We evaluate τ by fitting the autocorrelation function $C(t)$ of the end-to-end vector to an exponential function $C(t) = C_0 \exp(-t/\tau)$ [36]. The fitting range includes sufficiently long times, where higher-mode contributions are negligible for estimating τ . We show the results at different values of the Weissenberg number $Wi_\alpha = \tau\dot{\epsilon}_\alpha = 0.5, 1, 2$, and 6 . At $Wi_\alpha = 0.5$, $\eta_{E,p}^+(t;\dot{\epsilon})$, $\eta_{P,p}^+(t;\dot{\epsilon}_P)$, and $\eta_{B,p}^+(t;\dot{\epsilon}_B)$ almost overlap with the linear viscoelastic (LVE) envelopes $3\eta_{0,p}^+(t)$, $4\eta_{0,p}^+(t)$, and $6\eta_{0,p}^+(t)$, respectively. Here, we compute $\eta_{0,p}^+(t)$ as follows:

$$\eta_{0,p}^+(t) = \int_0^t G(t') dt' - \eta_s, \quad (12)$$

where $G(t)$ is the linear relaxation modulus obtained with the Green–Kubo formula. For $Wi_\alpha \gtrsim 1$, $\eta_{\alpha,p}^+(t;\dot{\epsilon}_\alpha)$ initially follows the LVE envelope, whereas it exhibits upward deviations at intermediate times, a phenomenon known as strain hardening, which has been experimentally observed in uniaxial extensional flows of polymer solutions [49–51]. As Wi_α increases, strain hardening occurs at shorter times and becomes more evident. Although strain hardening is observed in all the considered extensional flows, it is relatively weak in biaxial extensional flows. Specifically, at the steady state for a given $Wi_\alpha (\gtrsim 1)$, $\eta_{B,p}^+(t;\dot{\epsilon}_B)$ takes the smallest value, while $\eta_{E,p}^+(t;\dot{\epsilon}) \simeq \eta_{P,p}^+(t;\dot{\epsilon}_P)$. In the following, we aim to understand these behaviors of $\eta_{\alpha,p}^+(t;\dot{\epsilon}_\alpha)$ by focusing on the flow-kinematics dependence of the polymer conformation.

To relate the extensional viscosity growth function to the polymer conformation, we adopt an analysis method based

on a single-chain model, which is referred to as the Rouse-type model [52]. In the Rouse-type model, the original Rouse model [53] is extended to incorporate arbitrary interaction potentials between beads. Specifically, each bead follows the overdamped Langevin equation

$$\frac{d\mathbf{r}_i(t)}{dt} = (\nabla \mathbf{u})^\top \cdot \mathbf{r}_i(t) - \frac{1}{\zeta} \frac{\partial \mathcal{U}(\{\mathbf{r}_j(t)\})}{\partial \mathbf{r}_i(t)} + \sqrt{\frac{2k_B T}{\zeta}} \mathbf{w}_i(t), \quad (13)$$

where $\mathbf{r}_i(t)$ is the position of the i -th bead, ζ is the friction coefficient, $\mathcal{U}(\{\mathbf{r}_j(t)\})$ is the interaction potential for a single polymer chain, and $\mathbf{w}_i(t)$ is the Gaussian white-noise vector satisfying

$$\langle \mathbf{w}_i(t) \rangle = 0, \quad (14)$$

$$\langle \mathbf{w}_i(t) \mathbf{w}_j(s) \rangle = \delta_{ij} \delta(t-s) \mathbf{I}, \quad (15)$$

where δ_{ij} is the Kronecker delta, $\delta(t)$ is the Dirac delta function, and $\mathbf{I}$ is the identity tensor. Equation (13) can be viewed as a coarse-grained, approximate equation of motion for a single tagged polymer after eliminating the degrees of freedom of the surrounding solvent and other chains. For the Rouse-type model, a relation between the stress tensor $\boldsymbol{\sigma}(t)$ and the polymer gyration tensor $\mathbf{S}(t)$ is derived as follows [52]:

$$\frac{d\mathbf{S}(t)}{dt} - (\nabla \mathbf{u})^\top \cdot \mathbf{S}(t) - \mathbf{S}(t) \cdot (\nabla \mathbf{u}) = -\frac{2}{\rho_p \zeta} [\boldsymbol{\sigma}(t) - \boldsymbol{\sigma}_{eq}], \quad (16)$$

where ρ_p is the number density of the beads, $\boldsymbol{\sigma}_{eq}$ is the stress tensor at equilibrium and $\mathbf{S}(t)$ is defined as

$$\mathbf{S}(t) = \left\langle \frac{1}{N_p} \sum_{i=1}^{N_p} \{\mathbf{r}_i(t) - \mathbf{r}_G(t)\} \{\mathbf{r}_i(t) - \mathbf{r}_G(t)\} \right\rangle. \quad (17)$$

Here, N_p is the number of beads in a chain, and $\mathbf{r}_G(t)$ is the position of the center of mass of a chain. By substituting $\nabla \mathbf{u}$ defined by Eqs. (4)–(6) into Eq. (16), we obtain

$$\eta_{\alpha,p}^+(t;\dot{\epsilon}_\alpha) = \rho_p \zeta \left[R_{g,+}^2(t;\dot{\epsilon}_\alpha) + \kappa_\alpha R_{g,-}^2(t;\dot{\epsilon}_\alpha) - \frac{1}{2\dot{\epsilon}_\alpha} \frac{d}{dt} \left\{ R_{g,+}^2(t;\dot{\epsilon}_\alpha) - R_{g,-}^2(t;\dot{\epsilon}_\alpha) \right\} \right], \quad (18)$$

where κ_α represents the ratio of the magnitude of the compressional strain rate and the extensional strain rate (i.e., $\kappa_E = 1/2$, $\kappa_P = 1$, and $\kappa_B = 2$). For clarity, we denote the

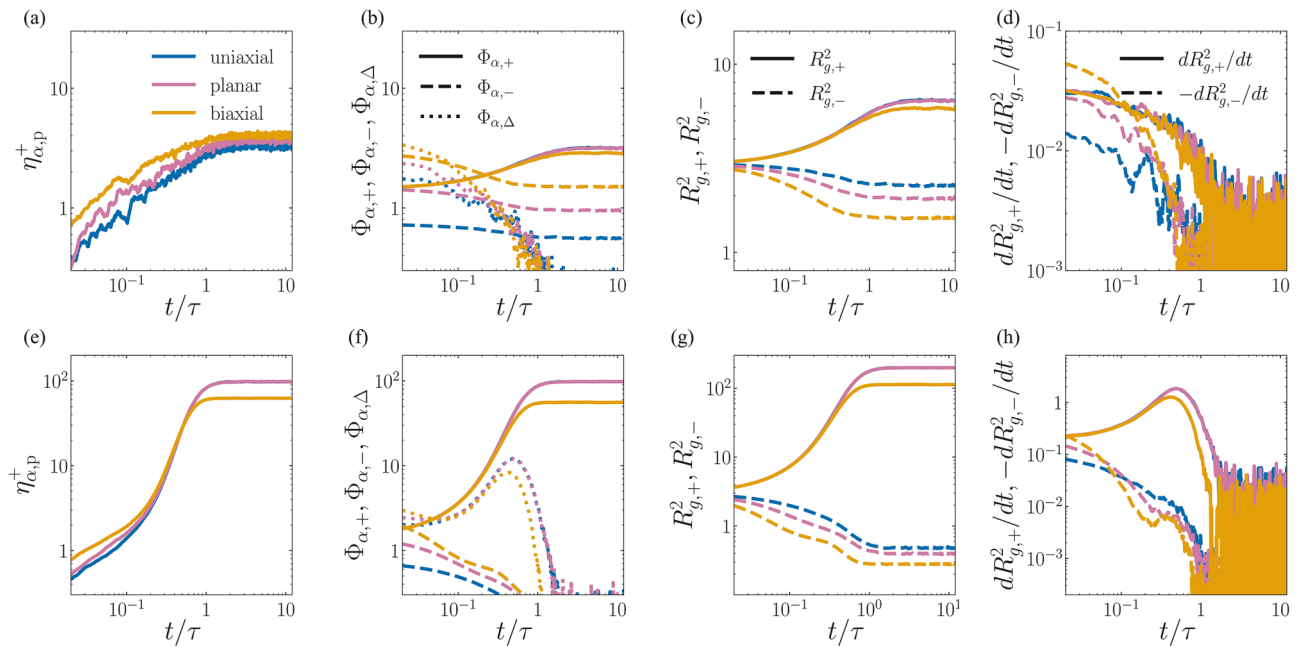


Fig. 4 Time series for **a–d** $Wi_\alpha = 1$ and **e–h** $Wi_\alpha = 6$: **a, e** $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$; **b, f** $\Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha)$, $\Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha)$, and $\Phi_{\alpha,\Delta}(t; \dot{\epsilon}_\alpha)$; **c, g** $R_{g,+}^2(t; \dot{\epsilon}_\alpha)$ and $R_{g,-}^2(t; \dot{\epsilon}_\alpha)$; **d, h** $dR_{g,+}^2(t; \dot{\epsilon}_\alpha)/dt$ and $-dR_{g,-}^2(t; \dot{\epsilon}_\alpha)/dt$

gyration radii in the extensional and compressional directions by $R_{g,+}(t; \dot{\epsilon}_\alpha)$ and $R_{g,-}(t; \dot{\epsilon}_\alpha)$, respectively. For instance, in biaxial extensional flows, $R_{g,+}(t; \dot{\epsilon}_B)$ and $R_{g,-}(t; \dot{\epsilon}_B)$ are defined as

$$R_{g,+}^2(t; \dot{\epsilon}_B) = \frac{1}{2} \left\langle \frac{1}{N_p} \sum_{i=1}^{N_p} [\{r_{i,x}(t) - r_{G,x}(t)\}^2 + \{r_{i,y}(t) - r_{G,y}(t)\}^2] \right\rangle, \quad (19)$$

$$R_{g,-}^2(t; \dot{\epsilon}_B) = \left\langle \frac{1}{N_p} \sum_{i=1}^{N_p} \{r_{i,z}(t) - r_{G,z}(t)\}^2 \right\rangle. \quad (20)$$

We explore whether the relation [Eq. (18)] obtained from the Rouse-type model is applicable to our systems. For this purpose, we estimate ζ , which is an a priori unknown parameter, by using the relation for the polymer contribution $\eta_{0,p}$ to the zero-shear viscosity η_0 in the Rouse-type model [52]:

$$\eta_{0,p} = \frac{\rho_p \zeta}{6} R_{g,eq}^2, \quad (21)$$

where $R_{g,eq}$ is the gyration radius at equilibrium. We calculate ζ by substituting $R_{g,eq}$ and $\eta_{0,p}$ obtained from the equilibrium DPD simulations into Eq. (21). In Fig. 3, we show the predictions based on the Rouse-type model computed from the right-hand side of Eq. (18) (black dashed curves). They are smoothed using the same moving average as $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$. These predictions quantitatively reproduce $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ obtained with the DPD simulations.

Although a slight discrepancy is observed at the steady state depending on Wi_α , which we discuss later in detail, we demonstrate that the Rouse-type model enables us to analyze $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ in terms of the polymer gyration radii $R_{g,+}(t; \dot{\epsilon}_\alpha)$ and $R_{g,-}(t; \dot{\epsilon}_\alpha)$. However, it should be noted that the applicability of Eq. (18) to our DPD simulations is nontrivial given that the Rouse-type model does not include hydrodynamic interactions. In our previous study [30], the manner in which ζ depended on N_p and ϕ suggested that the effects of hydrodynamic interactions may have been partly captured by ζ . Further investigations are needed to clarify why this framework works reasonably well, with a particular focus on the role of hydrodynamic interactions.

Utilizing the Rouse-type model, we explore how flow kinematics affect the extensional viscosity growth function in terms of the conformational changes of polymers. We focus on the contribution from each term on the right-hand side of Eq. (18). Specifically, we decompose $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ as $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha) = \Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha) + \Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha) - \Phi_{\alpha,\Delta}(t; \dot{\epsilon}_\alpha)$, where $\Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha)$, $\Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha)$, and $\Phi_{\alpha,\Delta}(t; \dot{\epsilon}_\alpha)$ are defined as

$$\Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha) = \rho_p \zeta R_{g,+}^2(t; \dot{\epsilon}_\alpha), \quad (22)$$

$$\Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha) = \kappa_\alpha \rho_p \zeta R_{g,-}^2(t; \dot{\epsilon}_\alpha), \quad (23)$$

$$\Phi_{\alpha,\Delta}(t; \dot{\epsilon}_\alpha) = \frac{\rho_p \zeta}{2 \dot{\epsilon}_\alpha} \frac{d}{dt} \{ R_{g,+}^2(t; \dot{\epsilon}_\alpha) - R_{g,-}^2(t; \dot{\epsilon}_\alpha) \}. \quad (24)$$

To investigate the contributions in different extensional flows at $Wi_\alpha = 1$, Fig. 4a shows $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$, and Fig. 4b shows $\Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha)$, $\Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha)$, and $\Phi_{\alpha,\Delta}(t; \dot{\epsilon}_\alpha)$. At $Wi_\alpha = 1$,

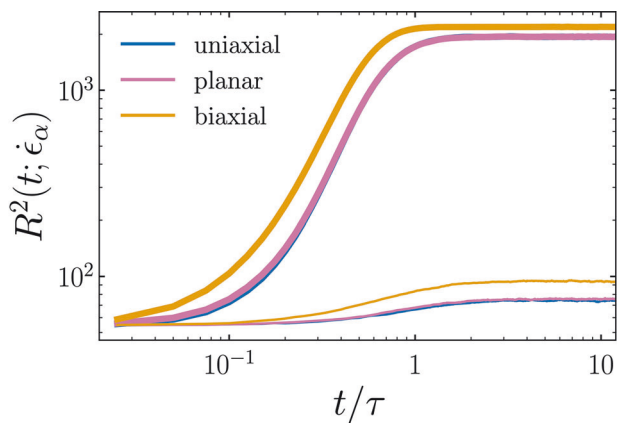


Fig. 5 Mean-square end-to-end distance $R^2(t; \dot{\epsilon}_\alpha)$ as a function of t/τ at $Wi_\alpha = 1$ (thin curves) and 6 (thick curves)

the relation $\eta_{B,p}^+(t; \dot{\epsilon}_B) > \eta_{P,p}^+(t; \dot{\epsilon}_P) > \eta_{E,p}^+(t; \dot{\epsilon})$ holds over the entire time range. In the growth regime of $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$, $\Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha)$, $\Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha)$, and $\Phi_{\alpha,\Delta}(t; \dot{\epsilon}_\alpha)$ exhibit comparable contributions to $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$. As t/τ increases, $\Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha)$ increases, $\Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha)$ decreases, and $\Phi_{\alpha,\Delta}(t; \dot{\epsilon}_\alpha)$ approaches 0. To relate these rheological responses to the polymer conformation, we also show $R_{g,+}^2(t; \dot{\epsilon}_\alpha)$ and $R_{g,-}^2(t; \dot{\epsilon}_\alpha)$ in Fig. 4c and $dR_{g,+}^2(t; \dot{\epsilon}_\alpha)/dt$ and $-dR_{g,-}^2(t; \dot{\epsilon}_\alpha)/dt$ in Fig. 4d. For $t/\tau \lesssim 1$, $R_{g,+}^2(t; \dot{\epsilon}_\alpha)$ is almost independent of the flow type because the strain rate in the extensional direction is common in these extensional flows, and the coupling between components is very weak. In contrast, $R_{g,-}^2(t; \dot{\epsilon}_\alpha)$ decreases most in the biaxial extensional flow and least in the uniaxial extensional flow, which is consistent with the magnitude of the compressional strain rate. Accordingly, $-dR_{g,-}^2(t; \dot{\epsilon}_\alpha)/dt$ takes the largest value in the biaxial extensional flow, whereas $dR_{g,+}^2(t; \dot{\epsilon}_\alpha)/dt$ is independent of the flow type. Although polymer compression reduces $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ (see Eq. (18)) and is strongest in the biaxial extensional flow, κ_α in Eq. (23) is also largest in this case. Thus, the relation $\eta_{B,p}^+(t; \dot{\epsilon}_B) > \eta_{P,p}^+(t; \dot{\epsilon}_P) > \eta_{E,p}^+(t; \dot{\epsilon})$ observed at $Wi_\alpha = 1$ mainly arises from the component $\Phi_{\alpha,-}(t; \dot{\epsilon}_\alpha)$ associated with the compressional direction, especially through κ_α , which is determined solely by the flow kinematics. Figure 4e–h shows the results at $Wi_\alpha = 6$. Although the initial response of $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ is similar to that at $Wi_\alpha = 1$, $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ in different extensional flows nearly collapse for $0.3 \lesssim t/\tau \lesssim 0.5$, and the relation $\eta_{E,p}^+(t; \dot{\epsilon}) \simeq \eta_{P,p}^+(t; \dot{\epsilon}_P) > \eta_{B,p}^+(t; \dot{\epsilon}_B)$ holds at the steady state [Fig. 4e]. Figure 4g demonstrates that at $Wi_\alpha = 6$, $R_{g,+}^2(t; \dot{\epsilon}_\alpha)$ becomes nearly two orders of magnitude larger than $R_{g,-}^2(t; \dot{\epsilon}_\alpha)$, thus leading to $\Phi_{\alpha,+}(t; \dot{\epsilon}_\alpha)$ being dominant in Fig. 4f. In addition, $R_{g,+}^2(t; \dot{\epsilon}_B)$ in the biaxial extensional flow becomes smaller than that in the other flows for $t/\tau \gtrsim 0.5$. Since the biaxial extensional flow has two extensional directions, the degree of stretching in each direction becomes smaller as polymers approach a fully extended state. Consequently, at $Wi_\alpha = 6$,

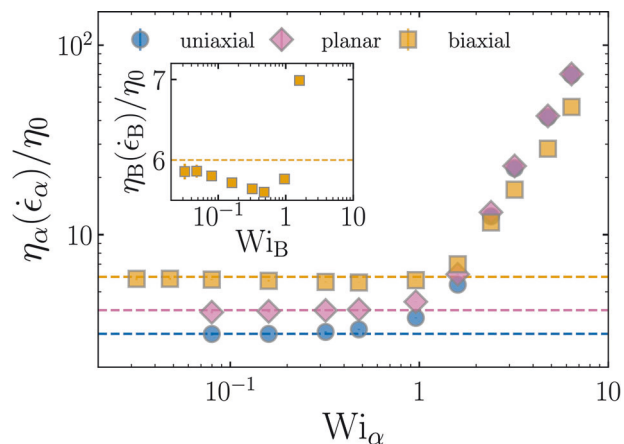


Fig. 6 Steady-state extensional viscosity $\eta_\alpha(\dot{\epsilon}_\alpha)$ normalized by the zero-shear viscosity η_0 as a function of the Weissenberg number Wi_α . The dashed lines indicate the corresponding Trouton ratios predicted by linear viscoelasticity: $\eta_E(\dot{\epsilon})/\eta_0 = 3$, $\eta_P(\dot{\epsilon}_P)/\eta_0 = 4$, and $\eta_B(\dot{\epsilon}_B)/\eta_0 = 6$. The inset shows a close-up view of $\eta_B(\dot{\epsilon}_B)/\eta_0$. The error bars denote the standard deviations from ten independent simulations

where nonlinear effects are nonnegligible, $\eta_{B,p}^+(t; \dot{\epsilon}_B)$ eventually takes the smallest value. Although $R_{g,+}^2(t; \dot{\epsilon}_\alpha)$, which is essentially related to $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$, becomes the smallest under the biaxial extensional flow, Fig. 5 demonstrates that the mean-square end-to-end distance $R^2(t; \dot{\epsilon}_\alpha) = \langle |\mathbf{r}_{N_p}(t) - \mathbf{r}_1(t)|^2 \rangle$ becomes the largest under the biaxial extensional flow at the considered Wi_α .

We then investigate the effects of flow kinematics on the steady-state extensional viscosity $\eta_\alpha(\dot{\epsilon}_\alpha)$ of dilute polymer solutions. We evaluate $\eta_\alpha(\dot{\epsilon}_\alpha)$ by time-averaging $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ over $t/\tau \gtrsim 10$. Figure 6 shows $\eta_\alpha(\dot{\epsilon}_\alpha)$ normalized by η_0 as a function of Wi_α . For $Wi_\alpha \ll 1$, $\eta_\alpha(\dot{\epsilon}_\alpha)/\eta_0$ approaches 3, 4, and 6 in the uniaxial, planar, and biaxial extensional flows, respectively, which is consistent with the Trouton ratios predicted by linear viscoelasticity [54]. For $Wi_\alpha \gtrsim 1$, $\eta_\alpha(\dot{\epsilon}_\alpha)$ increases with Wi_α , whereas $\eta_B(\dot{\epsilon}_B)$ slightly decreases for $0.1 \lesssim Wi_B \lesssim 1$ (see the inset of Fig. 6). In addition, as Wi_α increases, the relation $\eta_E(\dot{\epsilon}) \simeq \eta_P(\dot{\epsilon}_P) > \eta_B(\dot{\epsilon}_B)$ becomes apparent, which is consistent with recently reported experimental results [9]. The Weissenberg number at which $\eta_\alpha(\dot{\epsilon}_\alpha)$ begins to noticeably increase is also worth examining. For the FENE dumbbell model, $\eta_\alpha(\dot{\epsilon}_\alpha)$ sharply increases around $\tau_H \dot{\epsilon}_\alpha \simeq 0.5$ [55], where τ_H denotes the stress relaxation time in the Hookean limit. At first glance, this may appear inconsistent with our results, where $\eta_\alpha(\dot{\epsilon}_\alpha)$ begins to increase around $Wi_\alpha \simeq 1$. Note that the present study uses τ estimated from $C(t)$, rather than $G(t)$, because $G(t)$ exhibits significant statistical fluctuations in our simulations. This difference in the definition of the relaxation time partly explains the shift in the onset Weissenberg number, since in the Rouse model, the stress relaxation time is half the conformational relaxation time [56]. The limiting

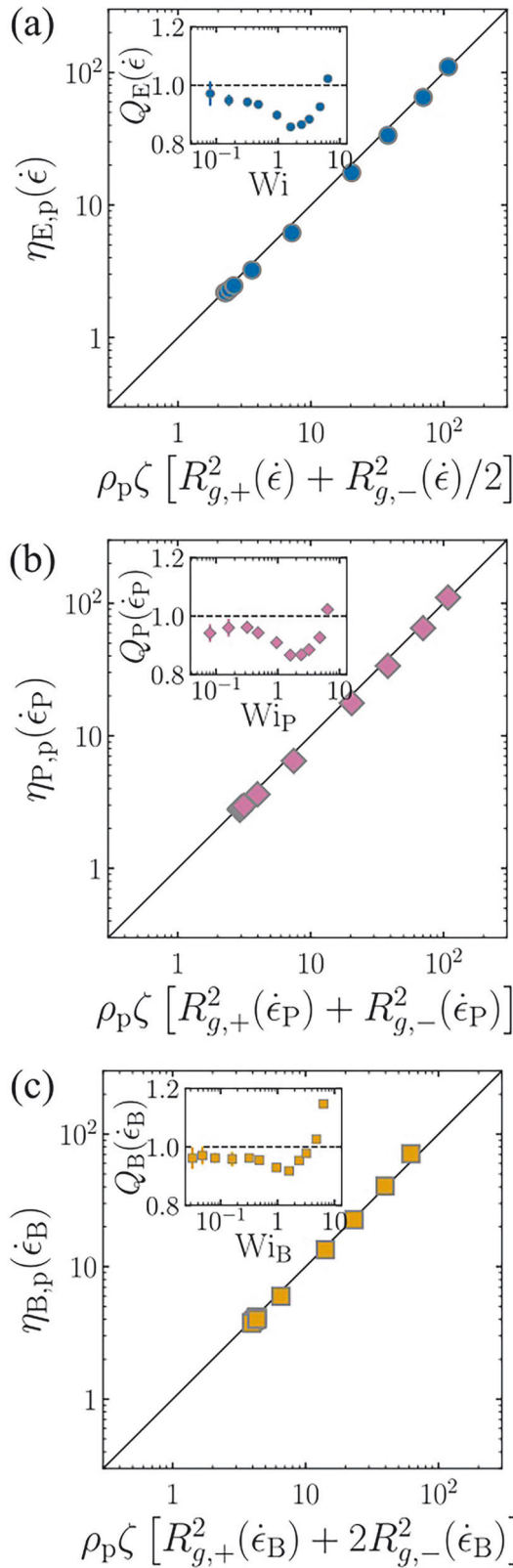


Fig. 7 Polymer contribution $\eta_{a,p}(\dot{\epsilon}_a)$ to the steady-state extensional viscosity $\eta_a(\dot{\epsilon}_a)$ as a function of $\rho_p \zeta [R_{g,+}^2(\dot{\epsilon}_a) + \kappa_a R_{g,-}^2(\dot{\epsilon}_a)]$ for **a** uniaxial, **b** planar, and **c** biaxial extensional flows. The black solid lines indicate $\eta_{a,p}(\dot{\epsilon}_a) = \rho_p \zeta [R_{g,+}^2(\dot{\epsilon}_a) + \kappa_a R_{g,-}^2(\dot{\epsilon}_a)]$. The insets in each panel show $Q_a(\dot{\epsilon}_a)$ as a function of Wi_a , where the black dashed lines indicate $Q_a(\dot{\epsilon}_a) = 1$. The error bars denote the standard deviations from ten independent simulations

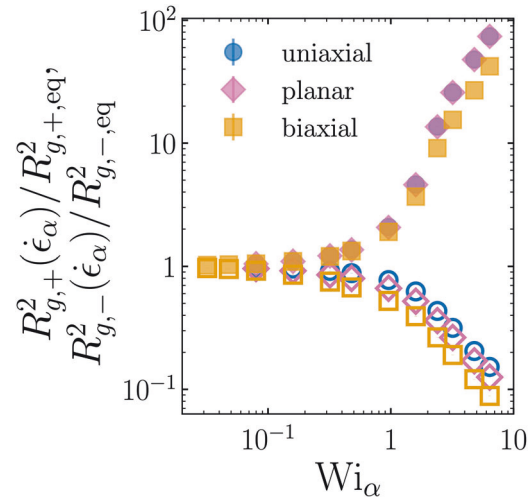


Fig. 8 Normalized mean-square gyration radii of the polymers in the extensional $R_{g,+}^2(\dot{\epsilon}_\alpha)/R_{g,+}^2(\dot{\epsilon}_{g,eq})$ (filled symbols) and compressional $R_{g,-}^2(\dot{\epsilon}_\alpha)/R_{g,-}^2(\dot{\epsilon}_{g,eq})$ (open symbols) directions as functions of the Weissenberg number Wi_α for uniaxial, planar, and biaxial extensional flows. The error bars denote the standard deviations from ten independent simulations and are smaller than the symbol size

are limited to $Wi_\alpha \lesssim 10$ due to the temperature increases observed at high extension rates (Fig. 2).

We aim to explain the flow-kinematics dependence of $\eta_a(\dot{\epsilon}_a)$ in terms of the polymer conformation. The Rouse-type model provides a quantitative relation between the steady-state extensional viscosity and the polymer gyration radius. At the steady state, Eq. (18) reduces to

$$\eta_{a,p}(\dot{\epsilon}_a) = \rho_p \zeta [R_{g,+}^2(\dot{\epsilon}_a) + \kappa_a R_{g,-}^2(\dot{\epsilon}_a)], \quad (25)$$

where $\eta_{a,p}(\dot{\epsilon}_a)$ is the polymer contribution to $\eta_a(\dot{\epsilon}_a)$. To verify the validity of Eq. (25), we show $\eta_{a,p}(\dot{\epsilon}_a)$ as a function of $\rho_p \zeta [R_{g,+}^2(\dot{\epsilon}_a) + \kappa_a R_{g,-}^2(\dot{\epsilon}_a)]$ in Fig. 7. For each extensional flow, $\eta_{a,p}(\dot{\epsilon}_a)$ is in good agreement with $\rho_p \zeta [R_{g,+}^2(\dot{\epsilon}_a) + \kappa_a R_{g,-}^2(\dot{\epsilon}_a)]$, including the results under the uniaxial extensional flow reported in our previous work [30]. For a more quantitative analysis, the inset of Fig. 7 shows the ratio $Q_a(\dot{\epsilon}_a)$ of the left-hand side to the right-hand side of Eq. (25), which is defined as $Q_a(\dot{\epsilon}_a) = \eta_{a,p}(\dot{\epsilon}_a) / \{\rho_p \zeta [R_{g,+}^2(\dot{\epsilon}_a) + \kappa_a R_{g,-}^2(\dot{\epsilon}_a)]\}$, as a

behavior of $\eta_a(\dot{\epsilon}_a)$ as $Wi_\alpha \rightarrow \infty$ is also important for comparisons with other models. However, the plateau region of $\eta_a(\dot{\epsilon}_a)$ is not reached in Fig. 6 because our results

function of Wi_α . Overall, Eq. (25) holds within $\pm 20\%$ irrespective of the flow type and Wi_α . However, $Q_\alpha(\dot{\epsilon}_\alpha)$ exhibits a nonmonotonic deviation from unity as Wi_α increases, with a weak dependence on the flow type. While we assume that the friction coefficient ζ is constant for the given N_p and ϕ values, the observed deviation of $Q_\alpha(\dot{\epsilon}_\alpha)$ from unity may be interpreted as the modulation of ζ due to extensional flows. Indeed, flow-induced changes in the effective friction coefficient have been extensively discussed as one of the key factors underlying the nonlinear responses under fast flows in both dilute [28, 57] and concentrated systems [58, 59]. Specifically, in dilute solutions, such changes have been attributed to the conformation dependence of hydrodynamic interactions [28, 57]. Refining our modeling approach to incorporate $\dot{\epsilon}_\alpha$ -dependent ζ is left for future work.

On the basis of the established relation (Fig. 7), we elucidate the physical origin of the flow-kinematics dependence of the steady-state extensional viscosity from the perspective of the polymer conformation. We first examine how the gyration radius depends on the flow type and the Weissenberg number. Figure 8 shows the mean-square gyration radii $R_{g,+}^2(\dot{\epsilon}_\alpha)$ and $R_{g,-}^2(\dot{\epsilon}_\alpha)$ in the extensional and compressional directions as functions of Wi_α . Here, both $R_{g,+}^2(\dot{\epsilon}_\alpha)$ and $R_{g,-}^2(\dot{\epsilon}_\alpha)$ are normalized by the corresponding equilibrium values $R_{g,+,\text{eq}}^2$ and $R_{g,-,\text{eq}}^2$, respectively, both of which are equal to $R_{g,\text{eq}}^2/3$. Although $R_{g,+}^2(\dot{\epsilon}_\alpha)$ is a monotonically increasing function of Wi_α regardless of the flow type, $R_{g,+}^2(\dot{\epsilon})$ and $R_{g,+}^2(\dot{\epsilon}_p)$ become larger than $R_{g,+}^2(\dot{\epsilon}_B)$ for $Wi_\alpha \gtrsim 1$. As discussed above, the strain rate in the extensional direction is the same for the three extensional flows at a fixed Wi_α , and the uniaxial and planar extensional flows have a single extensional direction, whereas the biaxial extensional flow has two extensional directions, thus leading to $R_{g,+}^2(\dot{\epsilon}) \simeq R_{g,+}^2(\dot{\epsilon}_p) > R_{g,+}^2(\dot{\epsilon}_B)$. In contrast, $R_{g,-}^2(\dot{\epsilon}_\alpha)$ is a monotonically decreasing function of Wi_α , and $R_{g,-}^2(\dot{\epsilon}_B) < R_{g,-}^2(\dot{\epsilon}_p) < R_{g,-}^2(\dot{\epsilon})$ holds according to the order of magnitude of the strain rate in the compressional direction. Combining these observations of the gyration radii and Eq. (25) allows us to explain the flow-kinematics dependence of $\eta_\alpha(\dot{\epsilon}_\alpha)$. For $Wi_\alpha \lesssim 1$, since $R_{g,+}^2(\dot{\epsilon}_\alpha)$ and $R_{g,-}^2(\dot{\epsilon}_\alpha)$ have comparable contributions because of the modest change in the polymer conformation, the relation $\eta_B(\dot{\epsilon}_B) > \eta_p(\dot{\epsilon}_p) > \eta_E(\dot{\epsilon})$ holds, where the difference arises mainly from the kinematic factor κ_α in Eq. (25). Notably, under the biaxial extensional flow, $\kappa_B = 2$, and $R_{g,-}^2(\dot{\epsilon}_B)$ decreases relatively rapidly, leading to a slight decrease in $\eta_B(\dot{\epsilon}_B)$ (i.e., $\eta_B(\dot{\epsilon}_B)/\eta_0 < 6$) for $0.1 \lesssim Wi_B \lesssim 1$. For $Wi_\alpha \gtrsim 1$, $R_{g,+}^2(\dot{\epsilon}_\alpha)$ becomes dominant because of significant polymer stretching, and $\eta_E(\dot{\epsilon}) \simeq \eta_p(\dot{\epsilon}_p) > \eta_B(\dot{\epsilon}_B)$ becomes evident owing to the difference in the degree of stretching in

the extensional direction. We therefore conclude that the flow-type dependence of the steady-state extensional viscosity arises from (i) the kinematic constant κ_α determined by the velocity gradient tensor and (ii) the flow-induced modulation of the gyration radii $R_{g,+}(\dot{\epsilon}_\alpha)$ and $R_{g,-}(\dot{\epsilon}_\alpha)$.

Conclusion

In this paper, we examined the responses of dilute polymer solutions under uniaxial, planar, and biaxial extensional flows using DPD simulations (Fig. 1). Dilute polymer solutions exhibited strain hardening during the start-up extensional flows at high extension rates, whereas the quantitative behaviors depended on the flow kinematics (Fig. 3). To elucidate the flow-kinematics dependence, we related the extensional viscosity growth function $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ to the polymer conformation using the Rouse-type model. We demonstrated that $\eta_{\alpha,p}^+(t; \dot{\epsilon}_\alpha)$ is quantitatively reproduced by the instantaneous gyration radii in the extensional ($R_{g,+}(t; \dot{\epsilon}_\alpha)$) and compressional ($R_{g,-}(t; \dot{\epsilon}_\alpha)$) directions, their time derivatives, and the flow-kinematics dependent constant κ_α , which reflects the ratio of compression to extension in the imposed flow field. Utilizing this relation, we explained the differences among the extensional viscosity growth functions of the three extensional flows in terms of purely kinematic effects and flow-induced polymer conformational changes (Fig. 4).

We also demonstrated the flow-kinematics dependence of the steady-state extensional viscosity $\eta_\alpha(\dot{\epsilon}_\alpha)$ (Fig. 6). At small Weissenberg numbers Wi_α , $\eta_E(\dot{\epsilon}) < \eta_p(\dot{\epsilon}_p) < \eta_B(\dot{\epsilon}_B)$. In contrast, as Wi_α increased, the relation $\eta_E(\dot{\epsilon}) \simeq \eta_p(\dot{\epsilon}_p) > \eta_B(\dot{\epsilon}_B)$ became gradually evident. The Rouse-type model also provided a relation between the polymer contribution $\eta_{\alpha,p}(\dot{\epsilon}_\alpha)$ to $\eta_\alpha(\dot{\epsilon}_\alpha)$ and the gyration radii in the extensional ($R_{g,+}(\dot{\epsilon}_\alpha)$) and compressional ($R_{g,-}(\dot{\epsilon}_\alpha)$) directions (Fig. 7). On the basis of this relation, we explained the flow-kinematics dependence of $\eta_\alpha(\dot{\epsilon}_\alpha)$ in terms of Wi_α -dependent changes in $R_{g,+}(\dot{\epsilon}_\alpha)$ and $R_{g,-}(\dot{\epsilon}_\alpha)$ (Fig. 8).

In the present study, we established a framework for analyzing the flow-kinematics dependence of the extensional viscosities of dilute polymer solutions. This framework, which relates extensional viscosities to polymer conformations, is expected to contribute to the understanding and control of complex flow phenomena from a molecular-level perspective, such as drag reduction in turbulent flows, where extensional viscosity plays a key role [60–62]. Another important direction for future research is to apply our approach to more complex systems, such as wormlike micellar solutions [39], which may also exhibit flow-kinematics-dependent scission and recombination kinetics.

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Compliance with ethical standards

Conflict of interest The authors declare no competing interests.

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