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Reduction of monomeric friction coefficient for linear isotactic polypropylene melts in very fast uniaxial extensional flow

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ABSTRACT

For the first time, the monomeric friction coefficient for fully aligned chains, $\zeta_{aligned}$, was determined for three linear isotactic polypropylene melts (iPP) using a high-strain-rate limiting value of uniaxial extensional viscosity, $\eta_{E,U,\infty}$, obtained from our recent experimental data [Drabek and Zatloukal, Phys. Fluids **32**(8), 083110 (2020)] and expression relating $\eta_{E,U,\infty}$ with $\zeta_{aligned}$, which was derived for a fully stretched Fraenkel chain [Ianniruberto *et al.*, Macromolecules **53**(13), 5023–5033 (2020)]. It was found that the obtained $\zeta_{aligned}$ value is lower by a factor of 2.9–5.0 (or even by a factor of 8.7–16.5 if the effect of polydispersity is included) compared to the equilibrium friction coefficient, ζ_{eq}, defined according to Doi and Edwards. This strongly supports recent arguments from rheological data and molecular simulations that a reduction in the friction coefficient must be considered in order to understand dynamics of polymer melts in very fast flows.

The current understanding of the dynamics of linear polymer melts in very fast flows is based mainly on nonlinear extensional data for nearly monodispersed polystyrenes (PS)[1–15] [exceptionally for polyisoprene (PI),[16] poly(n-butyl) acrylate (PnBA),[16] poly(methyl methacrylate) (PMMA),[4] and poly(tert-butylstyrene) (PtBS)[4,5]] and standard molecular theory based tube model predictions for linear, monodisperse polymers (considering reptation, chain-end fluctuations, thermal constraint release, orientation, retraction after chain stretch, convective constraint release, and finite extensible nonlinear elasticity).[1,3,17] Theory predicts a constant extensional viscosity at low strain rates (i.e., below the reciprocal value of reptation time, $1/\tau_d$) due to the equilibrium state of macromolecules (Region I) and then decrease in viscosity with a slope of -1 due to orientation at medium strain rates reaching its minimum at the inverse of the Rouse time, $1/\tau_R$ (Region II) above which the viscosity starts to increase due to chain stretch (Region III). At the highest strain rates, the maximum chain stretch is reached and viscosity becomes constant again, equal to $\eta_{E,U,\infty}$[1,3,17,18] (Region IV, see Fig. 1). Two discrepancies were identified when these tube model predictions were compared with the corresponding experimental data (compare Figs. 1 and 2). First, the slope -1 in Region II does not correspond to the experimentally observed value -0.5 (or even lower in the case of the PnBA melt).[3] Second, the thinning-to-thickening region was identified at $\dot{\varepsilon} \sim 1/\tau_R$ for the entangled PS solutions (and PI and PnBA melts) but not for the high molecular weight entangled PS melts, although the $\dot{\varepsilon} \gg 1/\tau_R$ (or alternatively, if the stretch Weissenberg number was $Wi_R = \dot{\varepsilon}\tau_R \gg 1$).[1] The first discrepancy was attributed to the increased number of entanglements in Region II[3] and the second with the stretch/orientation induced reduction in the monomeric friction coefficient.[1,3]

Another difficulty that limits the understanding of polymer melt behavior in fast extensional flows is related to the experimental limitation of generating high enough strain rates for a sufficient time (i.e., high Hencky strains) to achieve steady state conditions. Currently used conventional experimental techniques, such as Filament Stretching Rheometers (FSR), allow generating extensional viscosity data at sufficiently high Hencky strains (up to 7[19]) but at relatively low extensional strain rates (usually a maximum of 0.2–20 1/s[2,20]) which allowed investigating only a limited number of polymers with a very high Rouse time, τ_R, usually in the range of tenths to hundreds of seconds ($\tau_R = 29.7$–360 s at T = 130 °C for linear PSs,[1,2,4–15] $\tau_R = 37.7$s at T = 150 °C for PMMA,[4] $\tau_R = 306$ s at T = 175 °C for PtBS[4]). For such polymers, strong extensional flows can be generated (in terms of

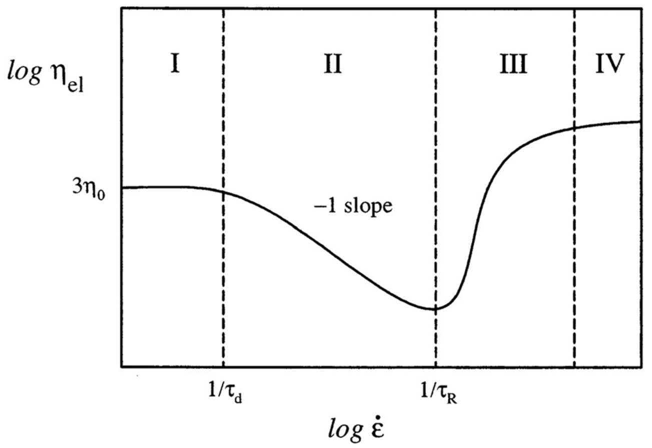

FIG. 1. Prediction of the steady-state uniaxial extensional viscosity vs extensional strain rate for linear, monodisperse polymer melt according to the standard tube model. Reproduced with permission from G. Marrucci, and G. Ianniruberto, "Interchain pressure effect in extensional flows of entangled polymer melts," Macromolecules **37**(10), 3934–3942 (2004). Copyright 2004 American Chemical Society.

the stretch Weissenberg number, i.e., when $\mathrm{Wi}_R > 1$), even if the applied strain rates are low. However, for polymers with a very low Rouse time, these techniques are not applicable due to the limitation of the maximum achievable strain rate. Very recently and for the first time, we have shown that it is possible to experimentally determine the extensional rheology of various PP melts at high Hencky strain (6.8) and extremely high strain rates ($2 \times 10^4 \sim 2 \times 10^6$ 1/s) utilizing entrance pressure drop measurements, an instrumented injection molding machine, and the Gibson method.[21–24] To fill the current knowledge gap with respect to the unknown dynamics of PP melts in fast extensional flows (the main aim of this work), our newly reported experimental data for linear iPP melts shown in Fig. 3 are analyzed here using the tube model with specific attention to the stretch induced reduction of the monomeric friction coefficient ζ.

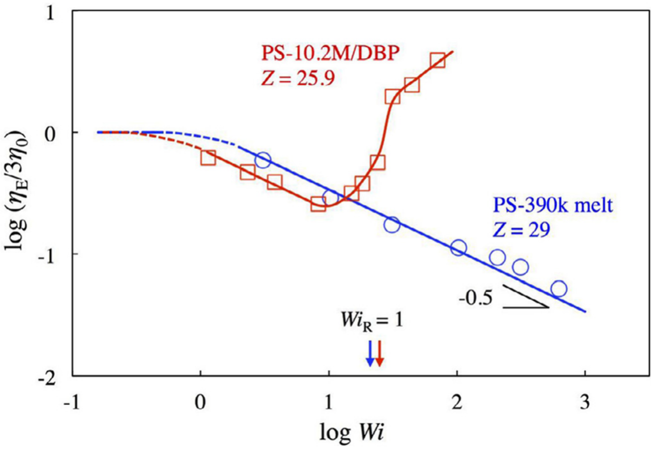

FIG. 2. Steady-state uniaxial extensional viscosity, $\eta_{E,U}$, normalized by the three times zero-shear rate viscosity, η_0, plotted as a function of the tube orientational Weissenberg number Wi ($= \dot{\varepsilon}\tau_d$) for PS solution (with dibutyl phthalate used as the solvent) at 21 °C and PS melt at 130 °C. Here, the location with the stretch Weissenberg number Wi_R ($= \dot{\varepsilon}\tau_R$) equal to 1 is provided by two arrows. Reproduced with permission from Y. Matsumiya and H. Watanabe "Non-universal features in uniaxially extensional rheology of linear polymer melts and concentrated solutions: A review," Prog. Polym. Sci. **112**, 101325 (2021). Copyright 2021 Elsevier.

Borflow HL504FB (iPP76k), HL508FB (iPP64k), and HL512FB (iPP56k) linear isotactic polypropylenes produced by Borealis Polyolefine (Linz, Austria) were used in this work. Their basic molecular characteristics are given in Tables I and II. The Rouse stress relaxation time is related to the equilibrium monomeric friction coefficient, ζ_{eq}, by the following Doi and Edwards expression:[17,25,26]

$$\tau_{R,s} = \frac{N^2 l^2}{6\pi^2 k_B T}\zeta_{eq}, \tag{1}$$

where l the Kuhn segment length, k_B the Boltzmann constant, T is the thermodynamic temperature, and N is the number of Kuhn segments

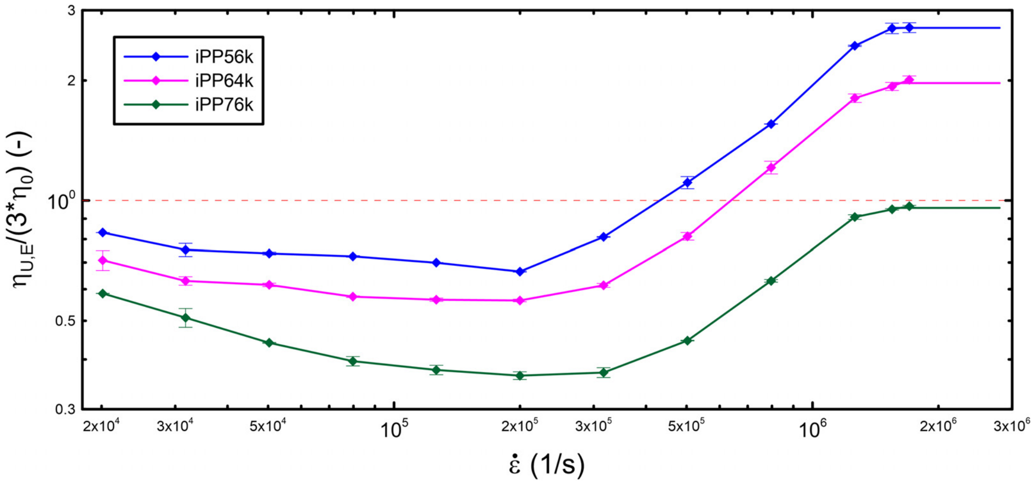

FIG. 3. Uniaxial extensional viscosity, $\eta_{E,U}$, normalized by the three times zero-shear rate viscosity, η_0, plotted as a function of extensional strain rate for linear isotactic polypropylene melts at 230 °C. Experimental data are taken from our previous work[24] and the lines should guide the eye.

TABLE I. Measured molecular and rheological characteristics for linear iPPs at T = 230 °C taken from Refs. 21–24.

	iPP76k	iPP64k	iPP56k
$\mathrm{M_n}$ (kg/mol)	17.20	14.650	14.25
$\mathrm{M_w}$ (kg/mol)	75.85	63.750	56.25
$\mathrm{M_z}$ (kg/mol)	165.50	138.00	114.50
$\mathrm{M_{z+1}}$ (kg/mol)	278.00	235.50	187.50
$\mathrm{IP = M_w/M_n}$	4.41	4.35	3.95
a	3.62	3.62	3.62
η_0 (Pa s)	22.80 ± 1.149	11.27 ± 0.671	7.79 ± 0.312
$\eta_{\mathrm{E,U},\infty}$ (Pa s)	65.56 ± 0.789[a]	66.62 ± 1.499[a]	63.32 ± 2.073[a]
$\eta_{\mathrm{E,U},\infty}/(3\eta_0)$	0.96 ± 0.012[a]	1.97 ± 0.044[a]	2.71 ± 0.089[a]

[a] These experimental values, which we extracted from our previous experimental data,[23,24] are reported here for the first time. Note that $\eta_{\mathrm{E,U},\infty}/(3\eta_0)$ values provided in Table II in Ref. 24 are predictions of the fitting equation [i.e., Eq. (12) in Ref. 24].

defined by the molar mass, M, and the Kuhn segment molar mass, M_k as

$$N = \frac{M}{M_k}. \tag{2}$$

The combination of Eqs. (1) and (2) leads to the following expression for the ζ_{eq}:

$$\zeta_{eq} = \frac{6\pi^2 k_B T}{l^2}\left(\frac{M_k}{M}\right)^2 \tau_{R,s}. \tag{3}$$

The Rouse stress relaxation time $\tau_{R,s}$ of linear entangled polymer melts can be determined using the following Osaki *et al.* formula[27] taking into account zero-shear rate viscosity dependence on the molar mass:

$$\tau_{R,s} = \frac{6M\eta_0}{\pi^2 \rho R T}\left(\frac{M_c}{M}\right)^{a-1}, \tag{4}$$

where M is the molar mass, η_0 the zero-shear rate viscosity, ρ the density, R the universal gas constant, T the thermodynamic temperature, a is the exponent in the range of 3.5±0.2,[17] and M_c is the critical molar mass at which entanglements starts to occur, which can be calculated according to Fetters *et al.*[28] as

$$M_c = M_e\left(\frac{p^*}{p}\right)^{0.65}. \tag{5}$$

Here, p is the packing length, which reflects the "degree to which molecules interpenetrate each other's space"[17] and $p^* = 9.2 \times 10^{-10}$m. M_e represents the molar mass between entanglements defined according to Ferry as $M_e = \frac{\rho RT}{G_N^0}$ with G_N^0 being the plateau modulus.[17]

TABLE II. Molecular characteristics for linear iPPs taken from Ref. 32.

ρ (kg/m^3)	766
$\mathrm{M_e}$ (kg/mol)	6.850
p (m)	3.12×10^{-10}
l (m)	11.4×10^{-10}
$\mathrm{M_k}$ (kg/mol)	0.1878
$\mathrm{M_c}$ (kg/mol)	13.935[a]

[a] The value was calculated according to Eq. (5).

Asymptotic expression for uniaxial extensional viscosity, $\eta_{E,U,\infty}$ (i.e., for $\dot{\varepsilon} \to \infty$), obtained for a Fraenkel chain incorporating finite chain extensibility, has been reported to be related to the friction coefficient for the fully aligned chain conformation, $\zeta_{aligned}$[3,29] (see also Refs. 30 and 31), as

$$\eta_{E,U,\infty} = \frac{\nu l^2 N^2}{12}\zeta_{aligned}, \tag{6}$$

from which

$$\zeta_{aligned} = \frac{12}{\nu l^2 N^2}\eta_{E,U,\infty}. \tag{7}$$

In these equations, the ν represents the Kuhn segment number density defined as

$$\nu = \frac{\rho N_a}{M_k}, \tag{8}$$

where N_a is the Avogadro number relating R and k_B as $\mathrm{N_a = R/k_B}$. Combination of Eqs. (2), (7), and (8) leads to the following final expression for the $\zeta_{aligned}$:

$$\zeta_{aligned} = \frac{12 k_B M_k^3 M^{-2}}{R \rho l^2}\eta_{E,U,\infty}. \tag{9}$$

ζ_{eq} and $\zeta_{aligned}$ were calculated for all three linear isotactic polypropylene melts at 230 °C according to Eqs. (3) and (9), respectively, using molecular constants (M_k, l, ρ, M_e, and p) taken from Fetters *et al.*[32] (see Table II) and experimental data ($M = M_w$, η_0, and $\eta_{E,U,\infty}$) taken from our recent experimental work[23,24] (see Table I). Here, it is assumed that the measured $\eta_{E,U,\infty}$ values correspond to the full-stretch state, i.e., the conditions for which Eq. (6) applies. The calculated values for the critical molecular weight, M_c, Osaki's Rouse stress relaxation time, $\tau_{R,s}$, the Rouse reorientation (or stretch) time, τ_R ($= 2\tau_{\mathrm{R,s}}$[17,33]) both friction coefficients and their ratio are given in Tables II and III considering $\mathrm{M = M_w}$. As can be seen from Table III, ζ_{eq} was found to be comparable for all three samples with an average value of $7.36 \pm 0.332 \times 10^{-12}$ Ns/m, which is the same order as reported in Ref. 34 for linear isotactic PP at 190 °C having $\mathrm{M = M_w}$ = 871 kg/mol and $\mathrm{M_w/M_n}$ = 2.4.[35] The $\zeta_{aligned}$ was found to be smaller compared to ζ_{eq}, and its value decreases with increased M_w as follows: 2.65×10^{-12} Ns/m for iPP56k, 2.17×10^{-12} Ns/m for iPP64k, and

TABLE III. Calculated molecular characteristics for linear iPPs at T = 230 °C considering $\mathrm{M = M_w}$.

	iPP76k	iPP64k	iPP56k
$\tau_{\mathrm{R,s}}$ (s)	3.87×10^{-6}	2.54×10^{-6}	2.15×10^{-6}
τ_{R} (s)	7.74×10^{-6}	5.08×10^{-6}	4.30×10^{-6}
ζ_{eq}(Ns/m)	7.51×10^{-12}	6.98×10^{-12}	7.59×10^{-12}
$\zeta_{aligned}$(Ns/m)	1.51×10^{-12}	2.17×10^{-12}	2.65×10^{-12}
$\zeta_{eq}/\zeta_{aligned}$	5.0	3.2	2.9

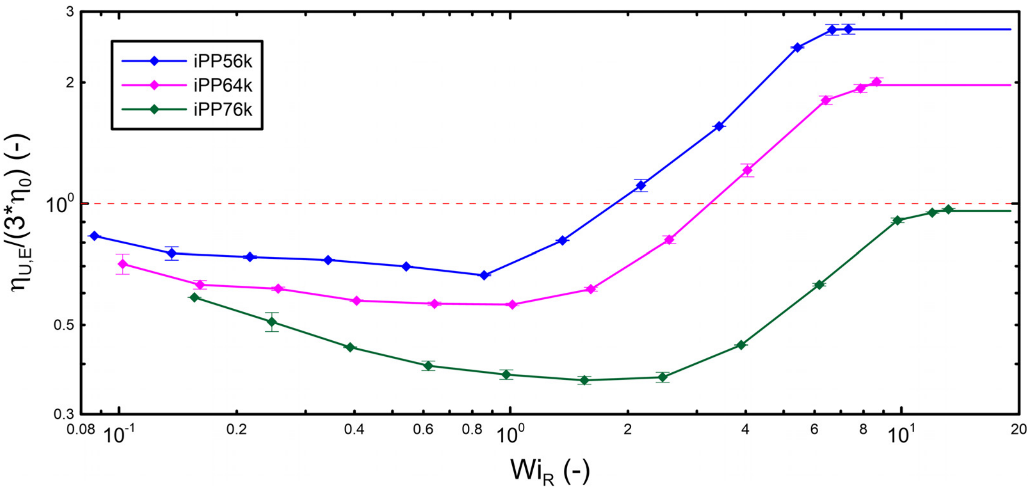

FIG. 4. Uniaxial extensional viscosity, $\eta_{E,U}$, normalized by the three times zero-shear rate viscosity, η_0, plotted as a function of stretch Weissenberg number ($= \dot{\varepsilon}\tau_R$) for linear isotactic polypropylene melts at 230 °C. Experimental data are taken from our previous work,[24] τ_R is given in Table III and the lines should guide the eye.

1.51×10^{-12} Ns/m for iPP76k. These values represent a reduction of ζ_{eq} by factor of 2.9 for iPP56k, 3.2 for iPP64k and 5.0 for iPP76k. It is interesting to note that the obtained reduction factors are between the theoretically predicted value of 2.6 for flexible chains[36] and 7.5 for chains with a fully aligned chemical environment.[29] The observed decrease in the ζ_{aligned} with the increased molar mass can thus be understood by the explanation provided by Park and Ianniruberto,[37,38] who showed that the reduction of ζ_{eq} can be promoted by nematic interactions between the molecular environment and polymer chains. They have shown that increasing the size of the surrounding chains allows "better connection" with the flow, leading to their co-alignment with the longest polymer chain, which reduces the friction coefficient.

Figure 4 shows uniaxial extensional viscosity data plotted as a function of chain stretch Weissenberg number, Wi_R. It can be seen that iPP56k and iPP64k samples with a low level of reduction in ζ show an extensional viscosity increase at $\mathrm{Wi_R} = 1$, which is consistent with the standard tube model predictions. However, in the case of the iPP76k sample, the transition from the thinning-to-thickening region is shifted to higher Wi_R values (about 2), which can be attributed to the observed high reduction in the ζ.

It is important to mention that all tested iPP melts (having $\mathrm{M_w} > \mathrm{M_c}$) are entangled in equilibrium (i.e., at very low strain rates), but they become fully disentangled at very fast flows.[21,22] In more detail, the secondary Newtonian viscosity, η_∞ (occurring above a shear rates of $2 \times 10^6\ \mathrm{s^{-1}}$) for all tested iPP melts was shown to be linearly dependent on the weight average molecular weight, M_w, confirming that the polymer chains became fully disentangled at the secondary Newtonian plateau region. This conclusion was further supported by the experimental observation that the high shear rate flow activation energy E_∞ for given iPP melts is comparable with the zero-shear rate flow activation energy, E_0, of PP like oligomer (squalane, $C_{30}H_{62}$; 2,6,10,15,19,23-hexamethyltetracosane with molecular weight equal to 0.4428 kg/mol). This justifies treating all tested iPPs in high stretch flow conditions as monodisperse unentangled polymer melts, in which the longest chains become stretched and the low molecular weight fractions create a specific chemical environment, which can take random or co-aligned orientation. In order to include the effect of the polydispersity index, $IP = \frac{M_w}{M_n}$, M can be expressed by M_n and M_w as follows:

$$M^2 = \sum_j x_j M_j^2 = \sum_j x_j M_j \left\{ \frac{\sum_j x_j M_j^2}{\sum_j x_j M_j} \right\} = M_n M_w, \tag{10}$$

from which

$$M = \sqrt{M_n M_w} \text{ or alternatively } M = \frac{M_w}{\sqrt{\mathrm{IP}}}. \tag{11}$$

Here, x_j is the number fraction of the chain having the molecular weight M_j. Assuming that M is given by $\frac{M_w}{\sqrt{\mathrm{IP}}}$, the key variables $\tau_{R,\ s}$, ζ_{eq}, and $\zeta_{aligned}$ calculated for $\mathrm{M} = \mathrm{M_w}$ increase by the multiplication factor $\mathrm{IP}^{\frac{a}{2}-1}$, $\mathrm{IP}^{\frac{a}{2}}$, and IP, respectively. In more detail, $\zeta_{eq}/\zeta_{\text{aligned}}$ is increased by the factor of $\mathrm{IP}^{\frac{a}{2}-1}$ to 8.7 for iPP76k, 10.6 for iPP64k, and 16.5 for iPP56k (see Table IV for more details). Interestingly, the maximum values of the $\zeta_{eq}/\zeta_{\text{aligned}}$ ratio for entangled and unentangled PS melt were reported to be approximately 10 and 100, respectively (see Fig. 9 in Ref. 1) Thus, it can be concluded that the obtained $\zeta_{eq}/\zeta_{\text{aligned}}$ ratio values between 2.9 and 5 (or 8.7 and 16.5 considering the polydispersity effect) for the tested iPPs are more comparable with the values for entangled PS melts than for unentangled PS melts.

TABLE IV. Calculated friction coefficients for linear iPPs at T = 230 °C considering $\mathrm{M} = \frac{M_w}{\sqrt{\mathrm{IP}}}$.

	iPP76k	iPP64k	iPP56k
ζ_{eq}(Ns/m)	1.10×10^{-10}	9.99×10^{-11}	9.12×10^{-11}
$\zeta_{aligned}$(Ns/m)	6.66×10^{-12}	9.44×10^{-12}	1.05×10^{-11}
$\zeta_{eq}/\zeta_{aligned}$	16.5	10.6	8.7

As the friction coefficient for the linear iPP melts control flow induced crystallization[34] and extensional rheology at high extensional rates, the results obtained are assumed to be valuable not only with regard to fundamental understanding of polymer physics and development/validation of advanced constitutive equations, but also with respect to understanding stability of non-Newtonian flows in advanced polymer processing, such as, for example, optimized production of polymer membranes for energy storage membranes[39,40] via cast film process[41] or filtration membranes prepared by the meltblown process[42] used, for example, in production of face masks.[43]

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DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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