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ORIGINAL PAPER

Correlation between entry pressure losses and elongation viscosity of polyethylene melts

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Abstract The correlation between the entry pressure drop and elongation viscosity during entry converging flow of polymer melts was discussed in this article. The entry pressure drop during extrusion of a low density polyethylene (LDPE) melt and a linear low density polyethylene (LLDPE) melt was measured by means of a capillary rheometer under test conditions with temperature of 170 °C and shear rate varying from 10 to 300 s^{-1}. The results showed that the entry pressure drop increased nonlinearly with an increase of the shear stain rate, and the variation of entry pressure drop of the two melts was close to each other. The melt elongation viscosity of the two resins was estimated using Cogswell equation from the measured entry pressure drop data, and the predictions were compared with the melt extension viscosity measured by using a melt spinning technique published in literature. It was found that the melt extension viscosity from entry converging flow was slightly lower than that from melt spinning technique under the same temperature and extension strain rate.

Keywords Polymer melt · Entry pressure losses · Elongation viscosity · Extrusion

Introduction

In polymer processing, entrance converging flow in die extrusion or runner injection of polymer melts from an extruder barrel is a usual flow pattern [1]. In general, the flow status of polymeric materials will affect significantly the processing technology and the final properties of products. Therefore, the melt extensional flow behavior

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and its mechanisms of polymeric materials have been paid attention by researchers in rheology and polymer processing engineers.

Entrance converging flow in die extrusion or runner injection of polymer fluids (including liquid and melt) has been studied extensively since 1970s [2–6]. Cogswell [2] studied the converging flow of polymer melts in extrusion dies, and proposed some simple methods for determining melt elongation stress and melt elongation viscosity. Binding [6] made an approximate analysis for contraction and converging flow, and derived an entry converging flow streamline equation. Liang [7] studied the converging flow during die extrusion of polymer materials, estimated the vortical region length of rubber compound during entry flow, and proposed an equation for determination of the entry region length of viscoelastic fluid flow in a channel [8]. Based on the smallest energy theory, Liang [9] derived an entry converging flow boundary streamline equation.

Entry convergent flow technique is an indirect method for measuring the extensional viscosity of polymer fluids. As stated above, entrance converging flow is a kind of elongation flow, there is a certain quantitative correlation between the entry pressure drop and extension viscosity, and hence the fluid extension viscosity may be calculated by measuring the entry pressure drop. Cogswell [2], Binding [6] and Gibson [10] proposed useful mathematical models to predict the melt extension viscosity from the entry converging flow of polymer melts. Comparing with direct method measuring extension viscosity such as melt spinning technique, the entry converging flow method is simpler and convenient.

LDPE and LLDPE are used extensively in plastics industry because of their good utility performance, processing properties and relative low cost. The objectives of this article are to study the relationship between the entry pressure losses and elongation viscosity during capillary extrusion flow and unique axial elongation flow of a LDPE and a LLDPE melts.

Theoretical description

Entry pressure losses

When a polymeric fluid (melt or liquid) enters a small channel from a reservoir, the streamline will not be parallel, and a conic boundary will be formed due to the viscoelasticity of the fluid and the contraction of the channel. This is an entry convergent flow, as shown in Fig. 1. Here, L_e is the circular flow zone length at front of the die, $2\alpha_0$ is the natural converging angle of fluid at the entrance, 2θ is the die angle, D_p and D are the diameters of the reservoir and die, respectively. It is generally believed that the entry convergent flow of polymer fluid consists of elongation flow and shear flow. In this case, the molecular chain will produce large extension deformation and shear deformation and relevant storage and viscous dissipation of the elastic deformation energy to form obvious entrance pressure losses. Pressure drop is a measurement of pressure losses. Therefore, the entry pressure drop (ΔP_{en}) include two parts: the pressure drop (ΔP_{ela}) generated from

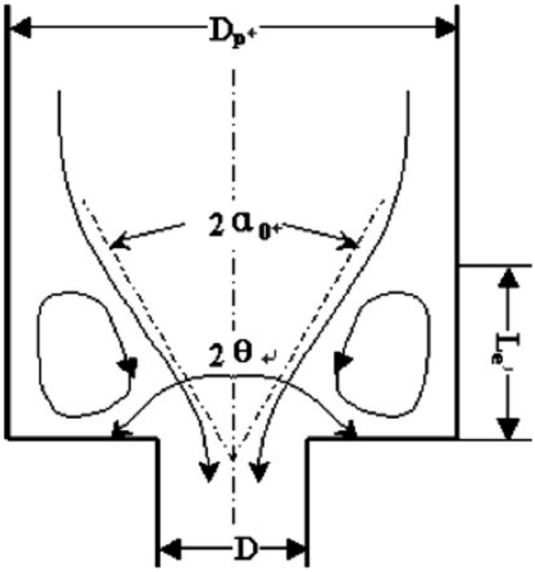

Fig. 1 Schematic of planar entry converging flow of polymer melt

the storage elastic deformation energy and the pressure drop (ΔP_{vis}) generated from the viscous dissipation of the elastic deformation energy. That is

$$\Delta P_{en} = \Delta P_{ela} + \Delta P_{vis} \tag{1}$$

In the previous work, the author [11] investigated the entry converging flow of polymer melt using tenser analysis method, and derived an expression of the ΔP_{ela} as follows:

$$\Delta P_{ela} = 2K \ln \lambda \left(2K_1 \dot{\gamma}_w^n + \frac{1}{2} K K_2 \dot{\gamma}_w^m \right) \tag{2}$$

While the viscous dissipation is given by:

$$\Delta P_{vis} = K_1 \dot{\gamma}_w^n \left[1 - (1/\lambda)^{3n} \right] / 3nK \tag{3}$$

Substituting Eqs. 2 and 3 into Eq. 1, we have

$$\Delta P_{en} = 2K \ln \lambda \left(2K_1 \dot{\gamma}_w^n + \frac{1}{2} K K_2 \dot{\gamma}_w^m \right) + K_1 \dot{\gamma}_w^n \left[1 - (1/\lambda)^{3n} \right] / 3nK \tag{4}$$

where λ is the channel contraction ratio, n and K_n, m, and K_m are, respectively, the power law constants of shear flow and elongation flow, $\dot{\gamma}_w$ is the shear rate, K is the entry converging flow parameter which is defined as follows:

$$K = \frac{1}{2} \tan \alpha_0 \tag{5}$$

where α_0 is the natural convergent angle of polymeric fluids (see Fig. 1).

Cogswell [2] analyzed the pressure losses when polymer melt flow through a conical die, and proposed an expression of the entry pressure drop on the basis of simplified hypothesis conditions:

$$\Delta P_{\mathrm{en}} = \frac{2}{3}\sigma_{\mathrm{e}}\left[1-\left(\frac{1}{\lambda}\right)^{3}\right] + \frac{2\tau_w}{3n\mathrm{tg}\alpha}\left[1-\left(\frac{1}{\lambda}\right)^{3n}\right] \tag{6}$$

where σ_{e} is the elongation stress, α is the converging angle of the die.

Elongation viscosity

As stated above, the entrance pressure losses are mainly due to the storage and viscous dissipation of the elastic deformation energy, while the elongation viscosity is related to the presence of viscous dissipation of the elastic deformation energy during entry converging flow of polymer melts. Hence there should be curtain correlation between the melt elongation viscosity and the entrance pressure losses. On the basis of the analysis on the polymer melt flow in conical die, Cogswell [2] proposed the following elongation viscosity (η_{el}) equation:

$$\eta_{\mathrm{el}} = \left[\frac{3\cdot(n+1)\cdot\Delta P_{\mathrm{en}}}{4\sqrt{2}\,\dot{\gamma}}\right]^{2}\cdot\frac{1}{\eta_{\mathrm{sh}}} \tag{7}$$

and the elongation strain rate is expressed as:

$$\dot{\varepsilon} = \frac{\dot{\gamma}}{2}\cdot\left[\frac{2\cdot\eta_{\mathrm{sh}}}{\eta_{\mathrm{el}}}\right]^{1/2} \tag{8}$$

where η_{sh} is the shear viscosity, which is given by:

$$\eta_{\mathrm{sh}} = \frac{\tau_w}{\dot{\gamma}_w} \tag{9}$$

and

$$\tau_w = \frac{(\Delta P - \Delta P_{\mathrm{en}})D}{4L} \tag{10}$$

$$\dot{\gamma}_w = \frac{32Q}{\pi D^3} \tag{11}$$

where Q is the volume flow rate.

Liang [1, 9] studied the entrant convergent flow of non-Newtonian fluid using the minimum energy theory, and derived the entry convergence boundary streamline equation. In the case of no wall-slip, a simplified elongation viscosity equation is written as follows:

$$\eta_{\mathrm{el}} = \left\{\Delta P_{\mathrm{ent}}\Big/\left[2\dot{\gamma}\Big/\left(\frac{4n}{3n+1}\right)^{\frac{1}{2}}\frac{2}{3(n+1)}\left(1-\left(\frac{R}{R_P}\right)^{\frac{3(n+1)}{2}}\right)\right]\right\}^{2}\Big/(2\eta_{\mathrm{sh}}) \tag{12}$$

Experimental

Raw materials

LDPE: the LDPE resin with trade-mark of 951-000 was supplied by China Petroleum and Chemical Corporation, Maoming city, China. The density and melt flow rate of the LDPE were 918 kg/m^3 and 2.17 g/10 min, respectively.

LLDPE: the LLDPE resin with trade-mark of DFDA-7042 was also supplied by China Petroleum and Chemical Corporation, Maoming city, China. The density and melt flow rate of the LLDPE were 920 kg/m^3 and 2.0 g/10 min, respectively.

Instrument and methodology

The melt extrusion rheological measurements were carried out by means of a capillary rheometer (Rheologic 5000) supplied by Ceast SpA in Italy, a set of capillary dies was selected, which the diameter was 1 mm, and the ratios of length to diameter (L/D) were 10, 30, and 40, respectively.

The rheological properties of the LDPE melt and the LLDPE melt were measured under test conditions with temperature of 170 °C and shear rates varying from 10 to 300 s^1. The entry pressure drop was determined using Bagley plotting method.

Results and discussion

Bagley correction

Under the same test conditions, the total pressure drop (ΔP) was measured as polymer melt flows through different length-diameter ratio (L/D) capillary dies with the same diameter. Then plotting the total pressure drop against the length-diameter ratio, one may obtain a series of linear curves of ΔP-L/D at the same temperature and corresponding shear rate or shear stress. The intercept of the curve at vertical coordinate is the entry pressure drop. This is the Bagley correction method. Figure 2 is the sketch of Bagley correction during extrusion of the LDPE at 170 °C when shear rates are varying from 10 to 300 s^{-1}.

It can be seen from Fig. 2 that the relationship between the total pressure drop and the length-diameter ratio of the LDPE melt and shear rate is linear. That is, the value of ΔP increases linearly with an addition of L/D when the shear rate is fixed. Figure 3 shows the relationship between the total pressure drop and the length-diameter ratio of the LLDPE melt under 170 °C and shear rate varying from 10 to 300 s^{-1}. It may be observed that the ΔP increases roughly linearly with an addition of L/D as the shear rate is constant.

Relatively, the linearity of the Bagley correction curves of the LDPE melt is better than that of the LLDPE melt, especially at higher shear rate level. For instance, the ΔP-L/D verves of the LDPE melt still keep good linearity at shear rate of 300 s^{-1}. While for the LLDPE melt, the linearity of the ΔP-L/D verve become

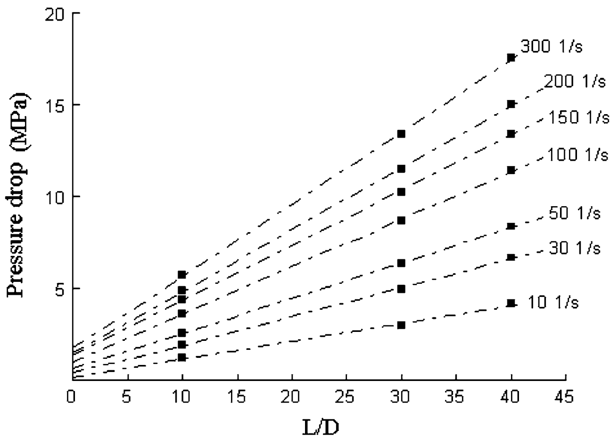

Fig. 2 Bagley correction of LDPE at 170 °C

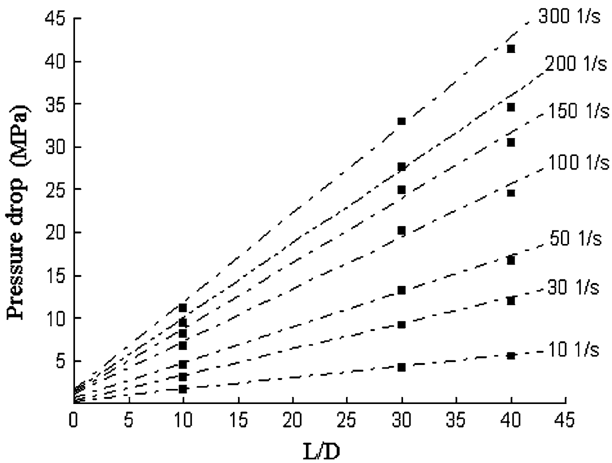

Fig. 3 Bagley correction of LLDPE at 170 °C

poor when shear rate is more than 50 s^{-1}. During extrusion of the LLDPE melt, the flow tends to unsteady at higher shear rates.

Entry pressure drop

Figure 4 displays the dependence of the entry pressure drop on shear rates for the LDPE melt and LLDPE melt at 170 °C. When shear rate is less than 150 s^{-1}, the entry pressure drop for the two resins melt increases quickly with increasing shear rate, then the entry pressure drop increases gently with an increase of shear rate. Moreover, the dependence of the entry pressure drop on shear rates of the LDPE melt is similar to the LLDPE melt under the same experimental conditions.

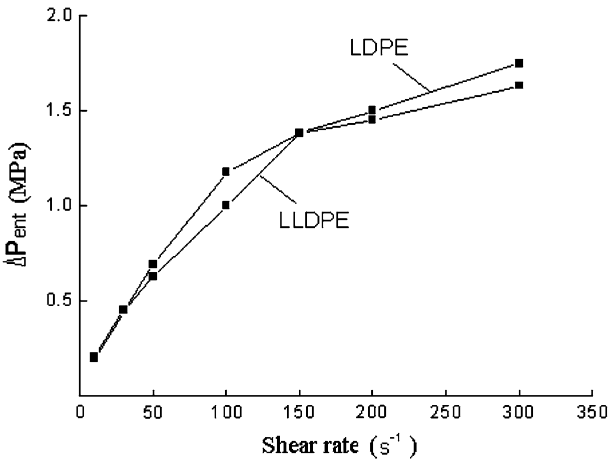

Fig. 4 Relationship between entry pressure drop and shear rate of LDPE and LLDPE at 170 °C

The macromolecular chains will be extended to start orientation along the flow direction during entry converging flow of polymer melts. In this case, a lot of storage and viscous dissipation of the elastic deformation energy will be generated, leading to obvious entry pressure losses. When the orientation of macromolecular chains is up to certain level, the variation will tend to slightly with the further increase of flow rate, while the flow resistance will decrease relevantly. In this case, the storage and viscous dissipation of the elastic deformation energy will increase slightly with increasing flow rate, resulting in increasing somewhat the entry pressure shear rate.

Estimation of elongation viscosity

Substituting the entry pressure drop determined by means of the Bagley correction method into Eq. 7, one may estimates the melt elongation viscosity of the LDPE and LLDPE resins under the experimental conditions. Then the elongation strain rate is determined by using Eq. 8. Figure 5 illustrate the dependence of the melt elongation viscosity of the LDPE and LLDPE resins on elongation strain rate at 170 °C. In a bi-logarithm coordinate system, the melt elongation viscosity decreases with increasing elongation strain rate. Similarly, the variation of the melt elongation viscosity with elongation strain rate for the two resins is close to each other. Furthermore, the value of the melt elongation viscosity of the LDPE is higher than that of the LLDPE at the same elongation strain rate, especially at lower elongation strain rate.

LDPE is a resin with branched molecular chains, LLDPE is a resin with linear molecular chains. It may be observed in experiments that the circle flow region will be formed at the front of the die during planar entry converging flow of LDPE melt, while there is no the circle flow phenomenon for the latter [12, 13]. Because the melt will make circulation flow in the circle flow region, the extra energy will be consumed to generate relevant entry pressure losses, resulting from increase of the

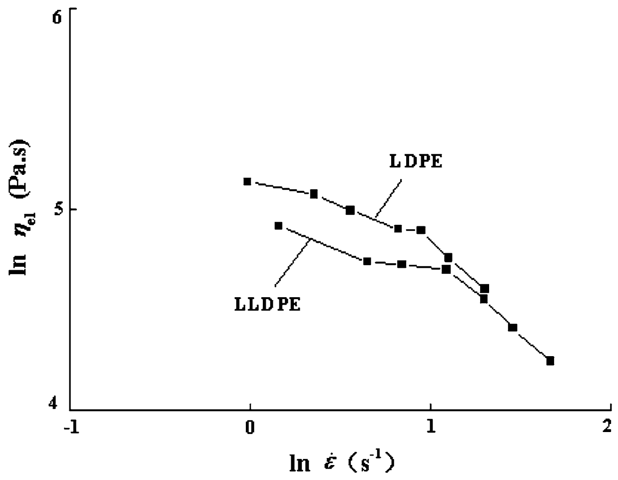

Fig. 5 Relationship between extensional viscosity and strain rate of LDPE and LLDPE at 170 °C

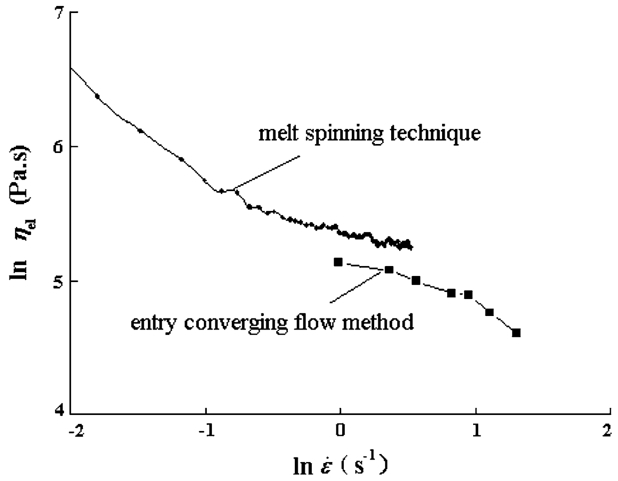

Fig. 6 Comparison of results of entrance flow and melt spinning of LDPE at 170 °C

melt viscosity. Hence the melt elongation viscosity of the LDPE is higher than that of the LLDPE at the same elongation strain rate (see Fig. 5).

Comparison and analysis

In the previous work, the authors [14] measured the melt elongation viscosity of the LDPE and LLDPE resins by means of a melt spinning technique at 170 °C. Figure 6 shows the comparison of the LDPE melt elongation viscosities between the measured from the entry converging flow data within elongation strain rate of 10^0–10^2 s^{-1} and measured using melt spinning technique within elongation strain rate of 10^{-2}–10^1 s^{-1}, the test temperature is 170 °C. It can be seen that the melt

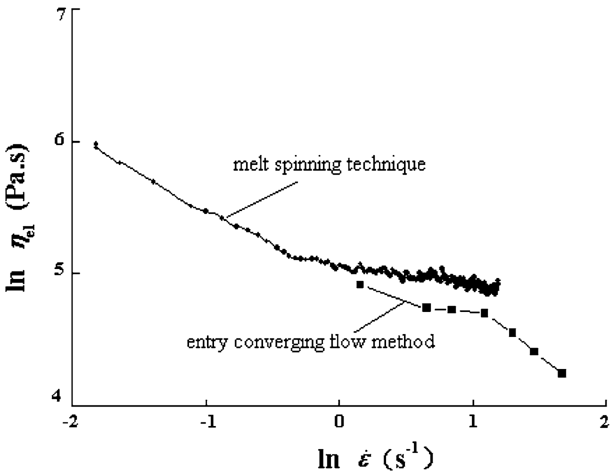

Fig. 7 Comparison of results of entrance flow and melt spinning of LLDPE at 170 °C

elongation viscosity decreases nonlinearly with an increase of elongation strain rate, and the values of the elongation viscosity measured using the melt spinning technique are slightly higher than those measured from the entry converging flow data at the same range of elongation strain rates.

Figure 7 shows the comparison of the LLDPE melt elongation viscosities between the measured from the entry converging flow data within elongation strain rate of 10^0–10^2 s^{-1} and measured using the melt spinning technique within elongation strain rate of 10^{-2}–10^1 s^{-1}, the test temperature is 170 °C. Similarly, the melt elongation viscosity decreases nonlinearly with an increase of elongation strain rate, and the values of the elongation viscosity measured using melt spinning technique are somewhat higher than those measured from the entry converging flow data at the same scope of elongation strain rates.

The factors of the difference in the melt elongation viscosity between the entry converging flow method and the melt spinning technique are complex. As stated above, the entry convergent flow of polymer fluid consists of elongation flow and shear flow while the melt spinning is a single axial elongation flow. Therefore, the values of the elongation viscosity measured using melt spinning technique are slightly higher than those measured from the entry converging flow data at the same scope of elongation strain rates. Thus, there will be somewhat difference in the measured data of the melt elongation viscosity between these two measurement methods. In other words, there is slight difference for the elongation viscosity values between measured by melt spinning technique and entry converging flow method (see Figs. 6 and 7).

Conclusions

The entry convergent flow consists of elongation flow and shear flow, and the entrance pressure losses are major due to the storage and viscous dissipation of the elastic deformation energy. The entry pressure drop during extrusion of a low density polyethylene (LDPE) melt and a linear low density polyethylene (LLDPE) melt was measured by means of a capillary rheometer under test conditions with temperature of 170 °C and shear rate varying from 10 to 300 s^{-1}. The results showed that the entry pressure drop increased nonlinearly with an increase of the shear stain rate, and the variation of entry pressure drop of the two melts was close to each other.

There is certain inherent correlation between the entry pressure losses and elongation viscosity during entry converging flow of polymer melts.The melt elongation viscosity of the two resins was estimated using Cogswell equation from the measured entry pressure drop data, and the predictions were compared with the melt extension viscosity measured by using a melt spinning technique published in literature. It was found that the melt elongation viscosity decreased with an increase of extension strain rate, and the melt extension viscosity from entry converging flow was slightly lower than that from melt spinning technique under the same temperature and extension strain rate.

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