Melt Strength, Local Velocity, and Elongational Viscosity Profiles of Low-Density Polyethylene Filaments Affected by the Die Design and Process Conditions

Watcharin Sitticharoen,[1] Wanlop Harnnarongchai,[1] Naret Intawong,[2] Narongrit Sombatsompop[1]

[1]*Polymer Processing and Flow Group, Division of Materials Technology, School of Energy, Environment and Materials, King Mongkut's University of Technology Thonburi, Thongkru, Bangmod, Bangkok 10140, Thailand*
[2]*Department of Industrial Engineering, Faculty of Engineering, Rajamangala University of Technology Lanna, 128 Huay Kaew Road, Chiang Mai, 50300, Thailand*

Received 22 March 2011; accepted 28 July 2011
DOI 10.1002/app.35378

ABSTRACT: An experimental arrangement to simultaneously measure the melt strength, velocity profiles, and elongational viscosity profiles across the cross section of a molten filament that emerged from either a circular or slit die for low-density polyethylene (LDPE) under nonisothermal and isothermal conditions is proposed. The proposed experimental rig was based on a parallel coextrusion technique of colored LDPE melt layers into an uncolored melt flowing from the barrel into and out of a die to form a continuous filament before they were pulled down by mechanical rollers until the filament failed. The experimental rig was also equipped with a high-speed data-logging system and a personal computer for real-time measurements. The results suggest that the draw-down forces changed continuously with changing roller speed, and the velocity profiles of the melt were not uniform across the LDPE filament during the stretching of the melt. Greater draw-down forces and local melt velocities were obtained in the slit die or under the nonisothermal condition. The draw-down forces and velocity profiles in both dies were affected by the volumetric flow rates from the extruder and the roller speeds used, with the effect being more pronounced for the circular die. The elongational viscosity profiles of the LDPE filament were not uniform across the filament cross section and corresponded well to the obtained velocity profiles. The elongational viscosities of the LDPE filament were relatively higher when the filament was extruded and stretched in the circular die and under the nonisothermal condition. The changes in the elongational viscosity profiles were more sensitive to changes in the volumetric flow rate and roller speed in the circular die. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 124: 3751–3764, 2012

Key words: mechanical properties; polymer extrusion; polyolefins; viscosity

INTRODUCTION

Melt spinning is a common process for generating molten polymer filaments and for measurements of melt properties under elongational deformation. The melt properties in this process are termed *melt strength*, *elongational stress*, and *strain* and *elongational viscosity*. Information on the elongational flow properties, extension force, and/or melt strength and melt viscosity is essential for understanding how the rheological behavior of polymer melts is greatly affected by the material properties (i.e., weight-average molecular weight, branching structure, and additives) and other process-related parameters, such as the volumetric flow rate, test temperature, roller speed and take-up style, draw ratio, ambient temperature, and die geometries.[1–11] Most polymers for elongational flow studies have been low-density polyethylene (LDPE),[4,8–10,11] linear low-density polyethylene (LLDPE),[4,7] high-density polyethylene,[1,3,4] polypropylene (PP),[2,4,5] and polystyrene.[4] Some polymer blends and composites have also been studied, including LLDPE/LDPE blends,[10,12–14] linear PP blends with long-chain-branched PP,[15] composite materials such as wood/thermoplastic composites,[16] wood/high-density polyethylene composites,[17] organoclay/polyamide 6 nanocomposites,[6] and carbon nanotube/polymers.[18] The die design and die temperature used for extruding the polymer melt also has a significant effect on the melt strength. Studies to improve the mechanical strength of polymer melts

---

Correspondence to: N. Sombatsompop (narongrit.som@kmutt.ac.th).

Contract grant sponsor: The Thailand Research Fund (The Thailand Research Fund Research Senior Scholar); contract grant number: RTA5280008.

Contract grant sponsor: Rajamangala University of Technology Lanna.

Contract grant sponsor: National Research University program by the Office of Higher Education Commission (OHEC) of Thailand.

Journal of Applied Polymer Science, Vol. 124, 3751–3764 (2012)

by materials modification[3,10–18] and die design.[2,5] have also been proposed. Gupta and Bhattacharya[2] clearly suggested that the melt strength of PP increased with increasing die diameter, whereas the opposite effect was observed when the die length was increased. The extensional viscosity for PP under the nonisothermal condition was also found to be higher than that under the isothermal condition.[5]

One of the parameters that has been reported to affect the mechanical strength of polymer melts is the velocity gradient, which is usually determined under shear flow deformation.[19] The most widely used technique has been laser–Doppler velocimetry (LDV).[4,20,21] This technique is relatively accurate but very expensive, complicated to use, and not robust.[4,20–23] To measure the melt strength and velocity profiles of the melt under elongation deformation, a combination of at least two techniques is required. Wagner et al.[4] studied the local velocity and elongational viscosity of several polymer melts using LDV and Göttfert Rheotens and indicated that the local velocity distribution along the spin line increased to a concave form with increasing draw-down force. The reduction of the apparent elongational viscosity during the flow was due to a preshearing effect. Schneider et al.[20] investigated the velocity distribution along the axis of an LDPE melt strand extruded through an axisymmetric capillary die using the LDV technique. The results suggest that the strand velocity pull-down by a Rheotens did not increase linearly with increasing distance from the die exit. It was found that the acceleration of the strand increased monotonically, and the velocity near to the die exit decreased with the draw-down force used. A strain-hardening effect was observed for LDPE but not for LLDPE, and the velocity profiles generated in the channel of the slit die were parabolic in form, with the melt velocity being high in the middle of the slit channel and low near the die wall.[21] Kim et al.[24] studied the velocity profiles of poly(ethylene terephthalate) filaments within the spinneret orifice and along the spin line using the finite element method and found that the melt velocity at the exit of the die channel was increased to a plug flow in form. Intawong and Sombatsompop[19,25] investigated the radial velocity profile of the polystyrene melt in shear flow using a parallel coextrusion technique (PCT). The results indicate that the velocity profile generated in the capillary die was parabolic in shape. The radial velocity profiles of the melt changed continuously with extrusion time.

On the basis of the work by Intawong and Sombatsompop,[19,25] the velocities of the melt across the flow channel were different, although they would equalize as they exited the die. However, during free flow, the melt at the filament skin may encounter a cooling effect. This would probably cause a velocity gradient across the melt. If this was the case, the elongational strain would be an uneven elongational strain and elongational viscosity across the extruded filament. However, these kinds of information have not yet been fully discussed and understood, and this has become our interest.

To measure the local elongation viscosity profiles of the LDPE melt under elongational deformation, the draw-down forces and local velocity profiles across the cross section of the LDPE extruded filament during the spinning process had to be measured accurately. This study was carried out with an experimental rig that was specially designed and originally developed on the basis of PCT[19] to simultaneously measure the local velocity profiles and the melt strength of the LDPE. Two different die designs were used, circular and slit dies, for comparison purposes. In this work, the draw-down force was used as an indicator of the mechanical strength for the LDPE filament, and a relationship between the local velocity profiles and the local elongational viscosity profiles across the cross sections of the LDPE filaments extruded from either the circular or slit die were established and compared under nonisothermal and isothermal conditions through the effects of the volumetric flow rate and roller speed.

EXPERIMENTAL

Proposed experimental design and concept

In this work, PCT[19] was used for simultaneous measurements of the local velocity profiles and melt strength of an LDPE filament during a free-flow process (a melt-spinning process). The local melt velocity profile measurement via PCT was based on the parallel coextrusion of colored LDPE molten rods (layers) into an uncolored LDPE melt stream from the barrel into and out of the capillary die. The melt velocity profiles at local positions across the die diameter were then measured by the introduction of relatively light and small particles into the melt layers, and the times taken for the particles to travel for a given distance were measured. The melt strength in this study was referred to as the draw-down force measured during the stretching of the extruded LDPE. The draw-down force and the measured velocities measured across the cross section of the LDPE filament extruded from either the circular or slit die were then used to measure the elongational melt velocities across the filament cross section.

Raw materials

LDPE (LD1905FA), with a melt flow rate of 5 g/10 min, was used a skin layer and was supplied by Thai Polyethylene Co., Ltd. (Bangkok, Thailand). On the basis of our previous work,[25] a red master batch (3.0%, Clariant Co., Ltd., Bangkok, Thailand) was used effectively

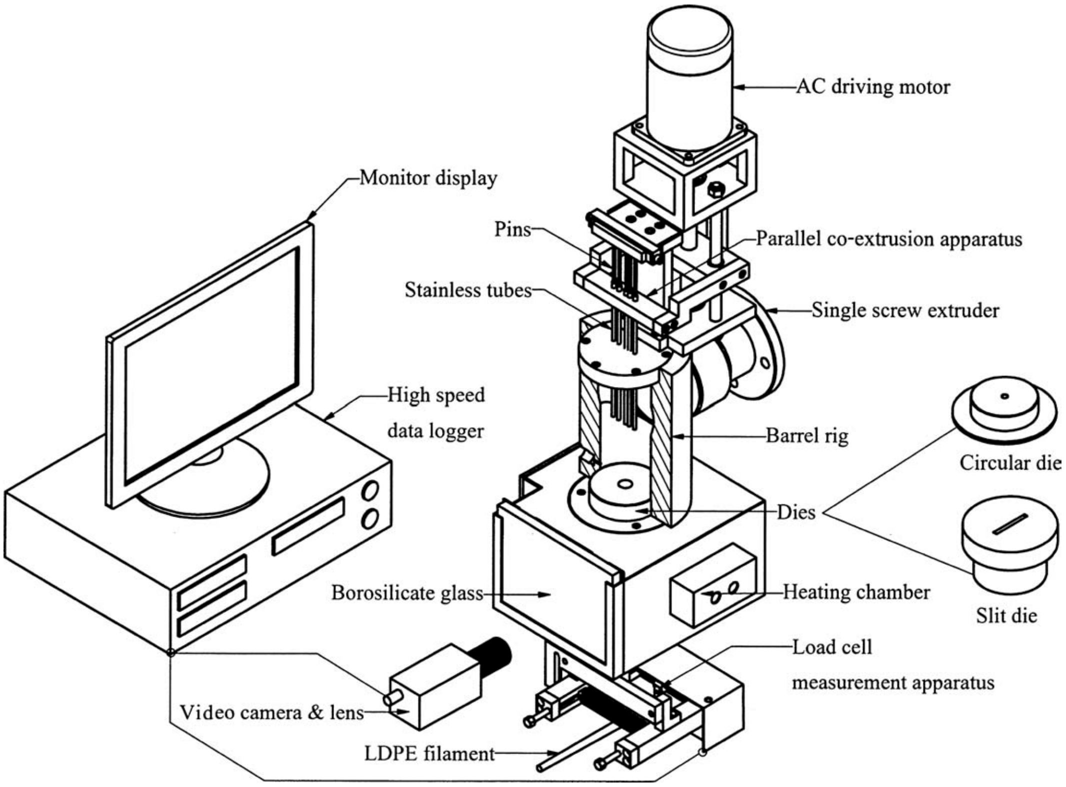

Figure 1 Experimental arrangement for the PCT for the measurement of the velocity profiles and melt strength of the LDPE filament; AC, alternating current.

for the colored LDPE core layers in the coextrusion process in the PCT. Corn particles (0.1 wt %) were used to follow the melt velocities in the colored LDPE core layer. The corn particles were relatively small, having an average particle size of 210 μm, and the maximum moisture content allowed was less than 5%.

Preparation of the LDPE colored layer rods

Neat LDPE and 3% red master batch were melt-blended in a single-screw extruder (En Mach Co., Ltd, Nonthaburi, Thailand) with a blending temperature profile on the extruder of 130, 140, 150, and 160°C from the hopper to die zones and a screw rotating speed of 10 rpm. A three-strand die, having a diameter of 3 mm for each strand and coupled with a palletizing unit, was used to produce the red LDPE pellets. The corn particles were dried to a constant weight to avoid any moisture during processing. Both the LDPE master-batched pellets and corn particles were held in an oven for 24 h at 80°C to avoid moisture before further processing. The experimental procedures to produce the red master-batch LDPE pellets with 0.1% corn particles can be found in our previous works.[19,25]

Experimental apparatus and procedures

PCT[19] coupled with roller take-up equipment, as given in Figure 1, was used for the measurements of the melt strength (as draw-down force) and the velocity profiles across the cross section of the LDPE filament. These two data sets could be used for the determination of the elongational viscosity profiles across the cross section of the LDPE filament. The experimental apparatus consisted of four parts: a single-screw extruder, an apparatus for measuring the melt strength, the PCT apparatus, and visualization equipment. The single-screw extruder (Thermo haake model PolyDrive, Karlsruhe, Germany) was used to produce the molten LDPE rods. The length-to-diameter ratio of the barrel was 450/19 mm/mm, and the temperature profile on the extruder from the hopper to the die zones was 130, 140, 150, and 160°C. The circular die used in this work was 20 mm in length and 6 mm in diameter, and the slit die was 30 mm in width, 3 mm in height, and 30 mm in length. The melt strength measuring apparatus consisted of two principal parts: a load cell and a roller take-up device. The force, measured with a load cell, was measured in the range 0–444 cN. The roller or take-up speeds from 0 to 120 m/min were used to wind the molten filament. The parallel coextrusion apparatus involved coextrusion of the colored layer of the LDPE melt in the stainless tubes into the uncolored melt flowing into the barrel when all of the stainless tubes moved upward by a moving arm. The details of this technique were discussed elsewhere.[19] The visualization equipment was used to

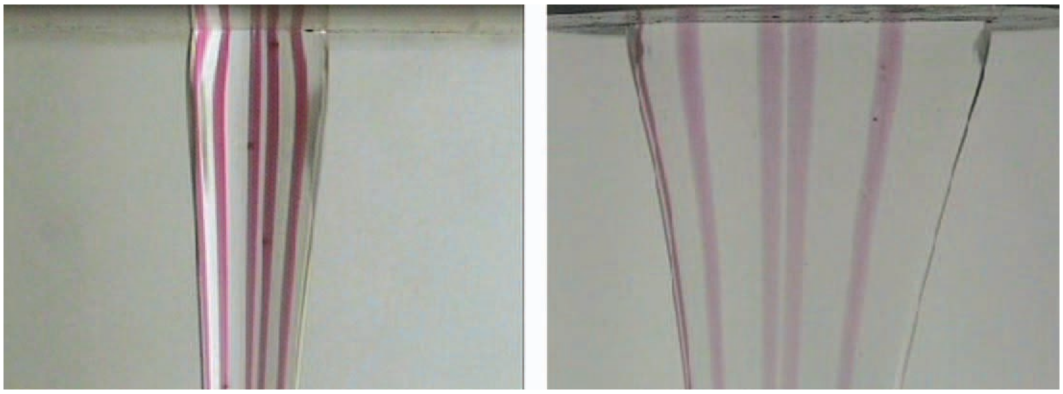

Figure 2 Flow visualization results for colored LDPE layers flowing in the LDPE filament at the die exit under stretching conditions: (a) circular die and (b) slit die. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

assist in monitoring the colored layers of the LDPE melt and the corn particles within the colored layers for the velocity profile measurements across the cross section of the LDPE filament. Figure 2 shows the flow visualization of the colored LDPE layers flowing within the LDPE filament with circular and slit dies during the stretching of the melt at the die exit region. Because this work used corn particles as foreign objects to follow the velocity of the LDPE melt and LDPE master batch for coextrusion, it was essential to determine whether the corn particles and the LDPE master batch affected the flow properties of the LDPE melt. Figure 3 shows the results of the apparent wall shear stress and apparent wall shear rate for the LDPE melts with and without additions of corn particles and the LDPE master batch at 160°C. It was found that the flow curves for both LDPE systems were very similar, with the differences being within an experimental error of ±4.9%. It was, therefore, confirmed that the corn particles and the master batch did not affect the flow properties of the molten LDPE.

In this work, the measurements of melt strength (as draw-down force), velocity profiles, and elongational viscosity profiles across the cross section of the LDPE filament were carried out under nonisothermal and isothermal conditions. The nonisothermal condition refers to the condition where the filament was cooled down by ambient air, and the isothermal condition refers to the condition in which the temperature of the filament was controlled by a heating chamber, which was made of stainless steel (grade SUS304) and the front of the chamber was made with borosilicate glass. The heating chamber used two infrared heaters (500 W, 220 V) and a DD6 temperature controller system. The temperature of the heating chamber was constant along the chamber length, with the differences being within an error of ±2.5°C.

Calculations of the local velocity profiles and local elongational viscosities

We carried out and monitored the velocity profile measurements by recording the times taken for the corn particles in the LDPE core layers to travel a given distance (25–50 mm from the die exit) in the LDPE filament. All of the experimental data were recorded and displayed with a high speed data-logging and recording system and a personal computer. The average melt velocity (v_n) of each colored layer across the cross section of the LDPE filament at the reduced radius (r/R) position for the circular die and reduced width (w/W) for a slit die could be calculated with eq. (1)

$$v_n = \frac{L_0}{t_c} \tag{1}$$

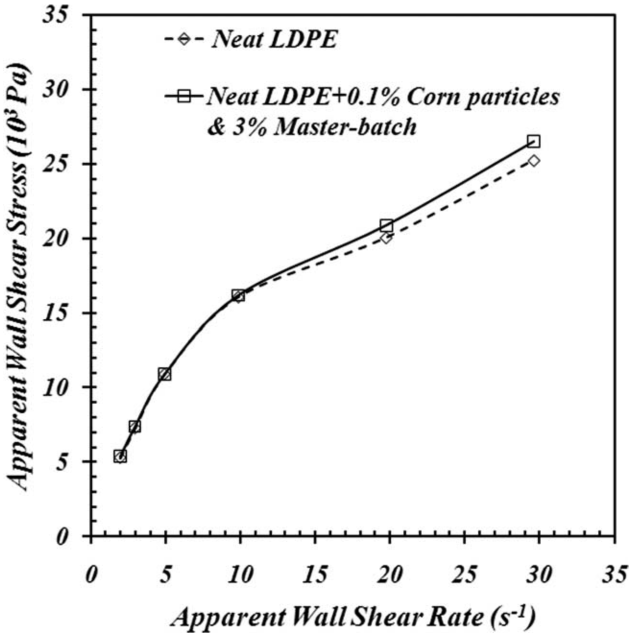

Figure 3 Flow curves for the molten LDPE with and without the corn particles and the LDPE master batch at a die temperature of 160°C.

TABLE I
Temperature Profiles under the Nonisothermal and Isothermal Conditions for the Circular Die at a Die Temperature of 160°C

Distance from the die exit (mm)	Temperature profile (°C)	
	Nonisothermal	Isothermal
5	120	160
25	107	162
50	84	161
60	76	156
75	67	137

where L_0 is the observed length of the measurement of 25×10^{-3} m for the circular die and 30×10^{-3} m for the slit die and t_c is the time that corn particle travels through L_0. Thus, the elongational viscosity (λ) is expressed by eq. (2):[1,6]

$$\lambda = \frac{FL}{Q\varepsilon_E} \tag{2}$$

where F is the average draw-down force with increasing step-ladder roller speeds, L is the length of the spin line, and ε_E is the elongational strain, which is defined as $\ln(\nu_n/\nu_0)$, and ν_0 is the melt velocity at the die exit. The viscosities for the LDPE melt were calculated with the melt velocities at any point across the filament diameters. The local elongational viscosities (λ_n) were calculated with the melt velocities, and the volumetric flow rate (Q) at any radial point across the LDPE filament diameter for the circular die is expressed by eq. (3), where r is the radius of each colored layer at the r/R position across the filament diameter and dr is the differentiate radius of each colored layer at the r/R position across the LDPE filament diameter:

$$\lambda_n = \frac{FL}{(2\pi r dr v_n) \times \ln\left(\frac{v_n}{v_0}\right)} \tag{3}$$

The local elongational viscosities across the filament cross section for the slit die is given by eq. (4):

$$\lambda_n = \frac{FL}{(WHv_n) \times \ln\left(\frac{v_n}{v_0}\right)} \tag{4}$$

where W is the thickness of the colored melt layer at each w/W position and H is the thickness of the LDPE filament.

Test variables

In this work, the test variables of interest included the volumetric flow rate and roller speed. The volumetric flow rate was varied from 4.5×10^{-8} to 9.0×10^{-8} m^3/s for the circular die and from 2.0×10^{-7} to 2.7×10^{-7} m^3/s for the slit die. It should be noted that there were two main reasons we did not use the same volumetric flow rates for the two dies. First, the geometries and dimensions of these two dies were different and on the basis of the volumetric flow rate calculations in eqs. (3) and (4), the volumetric flow rates obtained were expected to be different, even though one used the same screw-rotating speed. Second, the volumetric flow rate used for each die was deliberately selected and used so that the coextrusion process for the velocity profile measurement in each die was clearly visualized and accurate. The lengths of the spin line were fixed at 310 mm for the circular die and 295 mm for the slit die. The test temperature for the circular and slit dies was fixed at 160°C. The temperature profiles of the LDPE melt along the spin line under nonisothermal and isothermal conditions for the circular and slit dies are given in Tables I and II, respectively. It should be noted that the interesting filament temperature profiles were the same as those used for the velocity profile measurements, which were 25–50 mm away from the die exit. All of the reported experimental data are averages of at least five independent determinations.

RESULTS AND DISCUSSION

The main aim of this work was to study the effect of die geometry design and spinning process parameters (volumetric flow rate, take-up or roller speed, and isothermal condition) on the local elongational viscosity profiles across the cross section of the LDPE filament in a single-screw extruder with two different die geometries. On the basis of eqs. (3) and (4), to measure such local elongational viscosity profiles of the melt in the screw extruder with circular and slit dies, respectively, the draw-down forces and local velocity profiles across the cross section of the LDPE filament during the melt-spinning process were experimentally required.

TABLE II
Temperature Profiles under the Nonisothermal and Isothermal Conditions for the Slit Die at a Die Temperature of 160°C

Distance from the die exit (mm)	Temperature profile (°C)	
	Nonisothermal	Isothermal
5	120	164
25	104	165
50	86	160
60	76	156
75	68	136

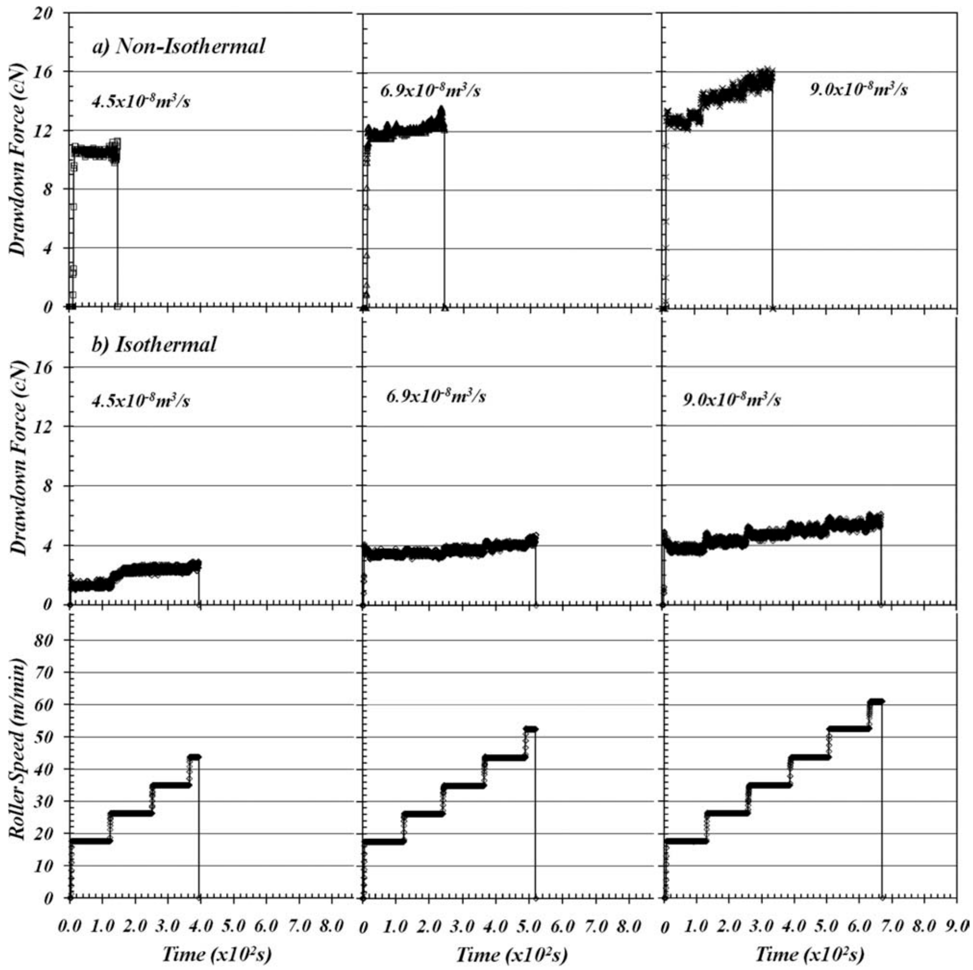

Figure 4 Draw-down force versus time as a function of the roller speed for the LDPE extrudate filament emerging from a circular die for three different volumetric flow rates: (a) nonisothermal filament stretching and (b) isothermal filament stretching.

Effects of the die geometry design and process parameters on the draw-down forces

Figures 4 and 5 show the draw-down force against extrusion time with increasing step-ladder roller speeds for the LDPE molten filament emerging from the circular and slit dies under nonisothermal and isothermal conditions for three different volumetric flow rates. The results indicate that the draw-down forces of the LDPE melt from the circular and slit dies sharply increased at the very beginning of the roller speed take-up and then gradually increased at further increasing roller speeds until the LDPE molten filament eventually failed. The changes in the draw-down forces with roller speed effect could be explained by the molecular entanglements and long-chain branching of LDPE, as detailed in our previous works.[8–10] For the effect of the volumetric flow rate from the extruder, it was found that the higher the volumetric flow rate was, the greater was the draw-down force that was required. The molten filament with a higher volumetric flow rate tended to generate greater stored energies in the molten polymer, and this enhanced the draw-down force. This claim was supported by the works of Baldi et al.[1] and Gupta and Bhattacharya[2] and our previous works.[8–10]

For the effect of die design, we found that the draw-down force of the LDPE molten filament from the circular die at any given volumetric flow rate under nonisothermal and isothermal conditions were lower than that in the slit die. This was because the output rates in the slit die were greater than those in the circular die at the same screw rotating speed from the extruder. However, the roller speed to failure of the molten filament from the circular and slit dies at any volumetric flow rate were the same. Comparing the results between Figures 4(a,b) and 5(a,b), we noticed that for any given roller speed, the draw-down forces for the molten filament from the circular and slit dies under the nonisothermal condition were greater than those for the isothermal condition. This was caused by the fact that the molten filament under the nonisothermal condition tended to solidify during filament

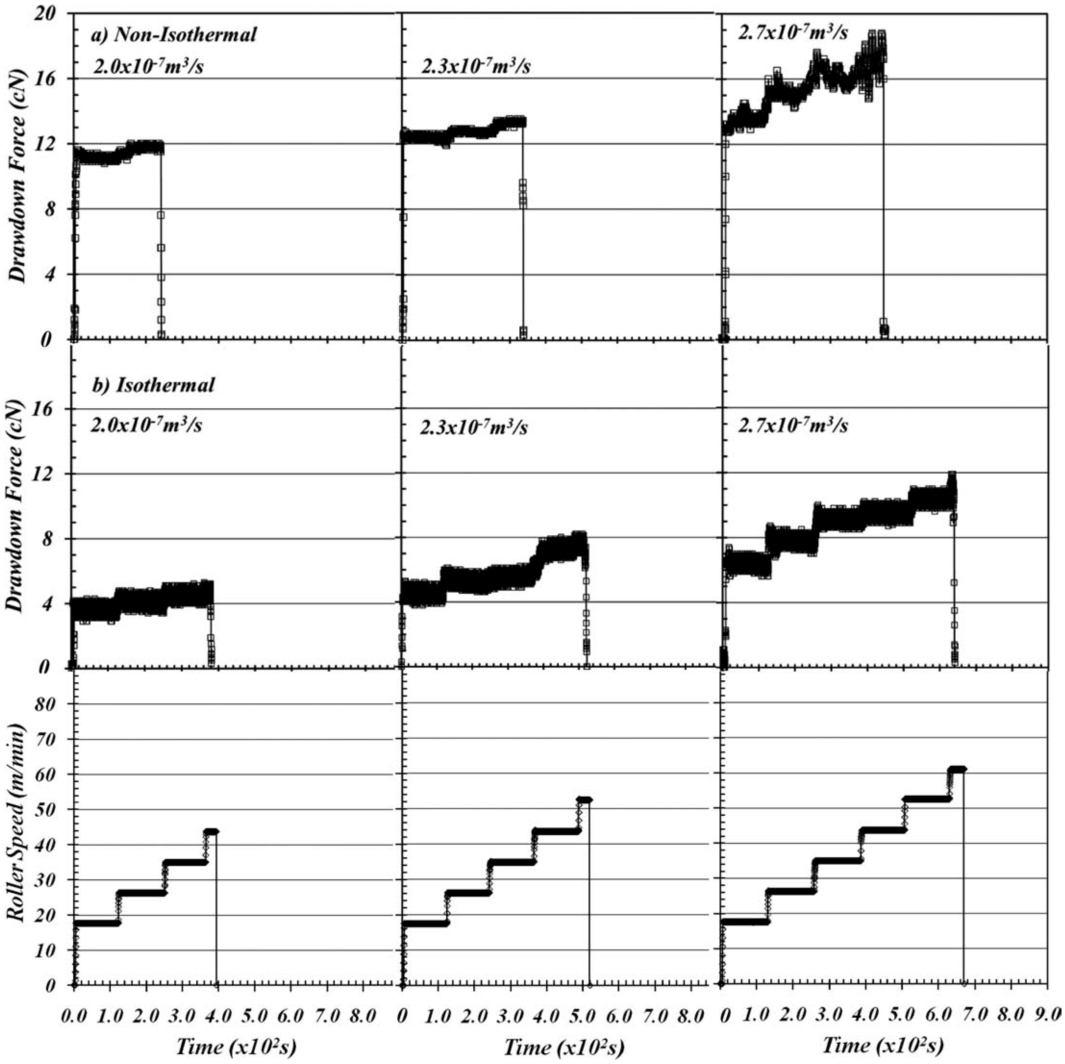

Figure 5 Draw-down force versus time as a function of the roller speed for the LDPE extrudate filament emerging from a slit die for three different volumetric flow rates: (a) nonisothermal filament stretching and (b) isothermal filament stretching.

stretching, especially on the surface of the molten filament, and this then led to a significant increase in the filament viscosity and, thus, increased drawdown forces. The elongational strains (or roller speed to failure) for the molten filaments from the circular and slit dies under the isothermal condition was greater than those under the nonisothermal condition. This may have been expected because the melt viscosity of the molten filament under the isothermal condition was lower and was not restricted by the cooling effect;[8,10,25] this allowed the molecular chains to slide past one another.

Effects of the die geometry design and process parameters on the local velocity profiles

Unstretched molten filament

Figure 6 shows the melt velocity profiles as a function of r/R position across the diameter for the LDPE molten filament under the nonisothermal and isothermal conditions without stretching by the rollers, and Figure 7 shows the velocity profiles as a function of w/W across the cross section for the LDPE molten filament under the nonisothermal and isothermal conditions without stretching by the rollers for three different volumetric flow rates. It was found that the velocity profiles of the molten filament from the circular die were pluglike, with the melt velocities across the filament diameters being very similar. This was expected, as the melt flowed in the die and exited the die lip; the explanations were given elsewhere.[24,25] The velocity profiles of the LDPE melt that emerged from the slit die were pluglike for the unstretched melt condition under the isothermal condition, but the velocities of the melt under the nonisothermal condition were relatively low near the edge of the filament strand. This was due to the cooling effect under the

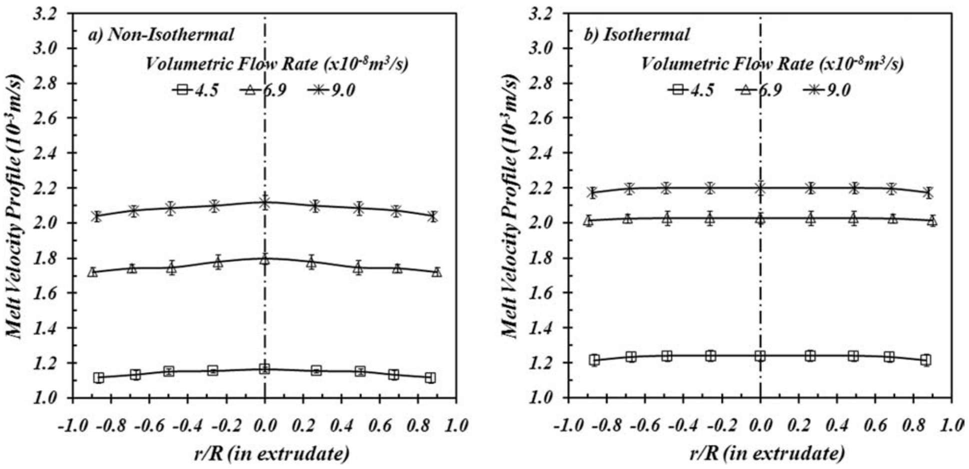

Figure 6 Melt velocity profiles for the LDPE filament emerging from a circular die without filament stretching for three different volumetric flow rates: (a) nonisothermal condition and (b) isothermal condition.

nonisothermal condition, as mentioned earlier. During free flow, the melt skin may have encountered cooler surroundings, and this would have probably caused the velocity gradient across the molten filament. However, the velocity gradients due to the cooling effect in the nonisothermal condition for the slit die was more pronounced than those for the circular die because of the relatively smaller width (3 mm) of the slit die as compared with the diameter (6 mm) of the circular die. The greater the volumetric flow rate from the extruder was, the higher the melt velocities were across the cross section of the molten filament for both die systems. For a given volumetric flow rate, the melt velocities for the molten filament under the isothermal and nonisothermal conditions for both die systems were slightly different, with the differences being within an experimental error of ±2.5%. However, the melt velocity profiles from the slit die under nonisothermal and isothermal conditions were greater than those of the circular die, again due to the fact that higher volumetric flow rates were obtained for any given screw speed from the extruder. In addition, during the experiment, the melt extruded from the circular die had greater swelling than that from the slit die; this indicated some transverse flow direction of the melt and/or a lower melt velocity in the axial flow direction. Greater swelling of the LDPE melt by the circular die relative to the slit die was also found in our previous work.[26]

Stretched molten filament

Figure 8 shows the melt velocity profiles as a function of r/R position across the diameter for the LDPE molten filament under nonisothermal filament stretching with increasing step-ladder roller speeds, and Figure 9 shows the melt velocity profiles as a function of w/W position across the cross section for the LDPE molten filament from the slit die under

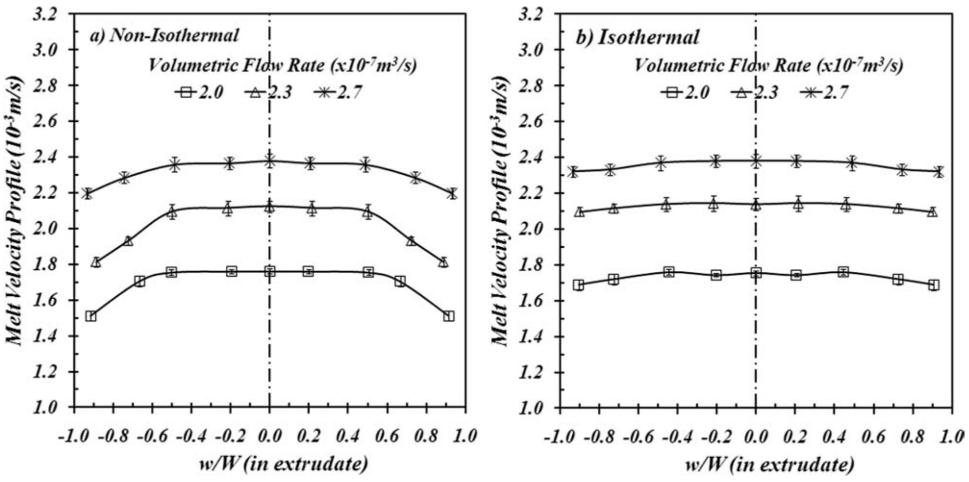

Figure 7 Melt velocity profiles for the LDPE filament emerging from a slit die without filament stretching for three different volumetric flow rates: (a) nonisothermal condition and (b) isothermal condition.

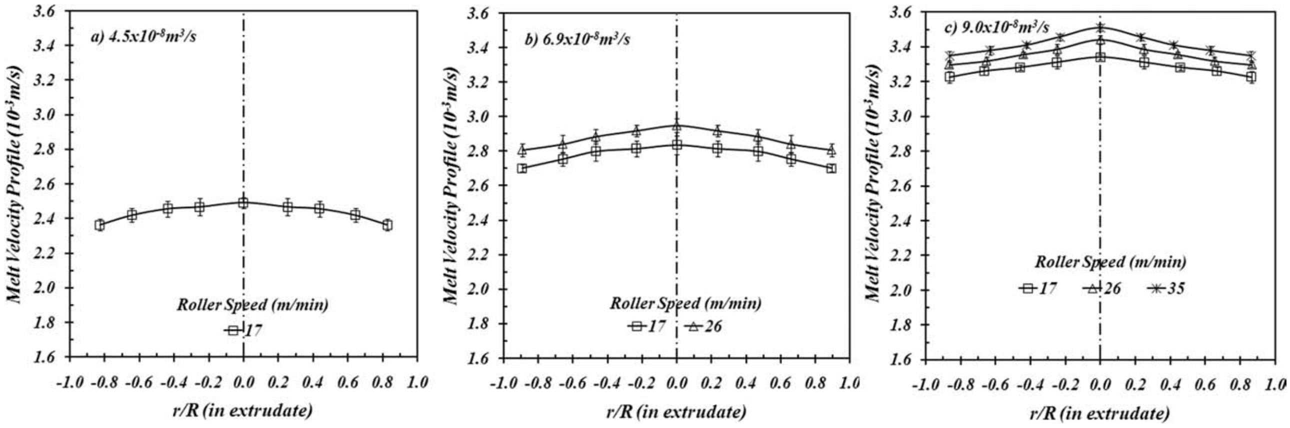

Figure 8 Melt velocity profiles for the LDPE filament from a circular die under nonisothermal filament stretching with increasing step-ladder roller speeds for three different volumetric flow rates: (a) 4.5 × 10^{-8}, (b) 6.9 × 10^{-8}, and (c) 9.0 × 10^{-8} m^3/s.

nonisothermal filament stretching with increasing step-ladder roller speeds for three different volumetric flow rates. It was observed that for a given volumetric flow rate, the velocity profiles of the molten polymer for both die systems decreased with increasing r/R position for the circular die and w/W position for the slit die, with a high melt velocity at the center position of the molten filament and a low melt velocity near the edge of the filament strand. The number of roller speeds used for each volumetric flow rates in the velocity profile measurements corresponded to those in the draw-down force measurements, as given in Figures 4 and 5. As the roller speed increased, the melt velocities at any radius/width point across the cross section of the molten filament increased. This view was supported by the work of Meerveld et al.,[27] who stated that the velocity profiles along spin line increased with increasing roller-speed take-up velocity. The melt velocities for both die systems also increased with increasing volumetric flow rate. It was interesting to note that the roller speed to failure for both die systems increased with increasing volumetric flow rate; that is, the filament at volumetric flow rates of 4.5 × 10^{-8} m^3/s for a circular die and 2.0 × 10^{-7} m^3/s for a slit die failed at a take-up speed of 26 m/min, whereas that at the volumetric flow rates of 9.0 × 10^{-8} m^3/s for a circular die and 2.7 × 10^{-7} m^3/s for a slit die failed at a take-up speed of 43 m/min. This was associated with the increased stored energies within the filaments during the extrusions with higher volumetric flow rates.

The results of local velocity profiles under the isothermal stretching condition for the circular and slit dies as a function of the volumetric flow rate and roller speed are given in Figures 10 and 11, respectively. The general patterns of the melt velocity profiles as a function of $r/R/w/W$ position, volumetric flow rate, and roller speed for both die systems were very similar to those in the nonisothermal stretching condition, as given in Figures 8 and 9. However, two differences were noted for both die

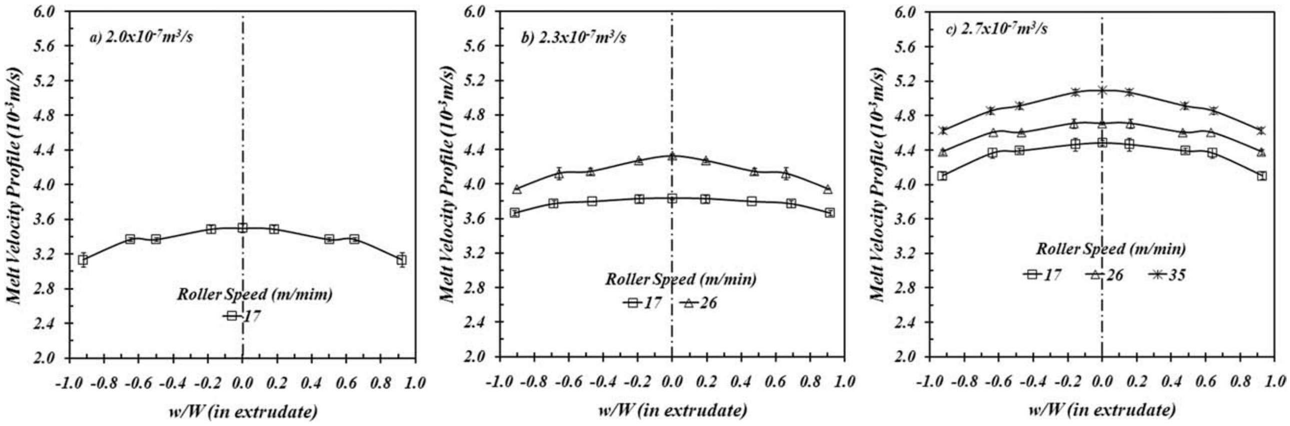

Figure 9 Melt velocity profiles for the LDPE filament from a slit die under nonisothermal filament stretching with increasing step-ladder roller speeds for three different volumetric flow rates: (a) 2.0 × 10^{-7}, (b) 2.3 × 10^{-7}, and (c) 2.7 × 10^{-7} m^3/s.

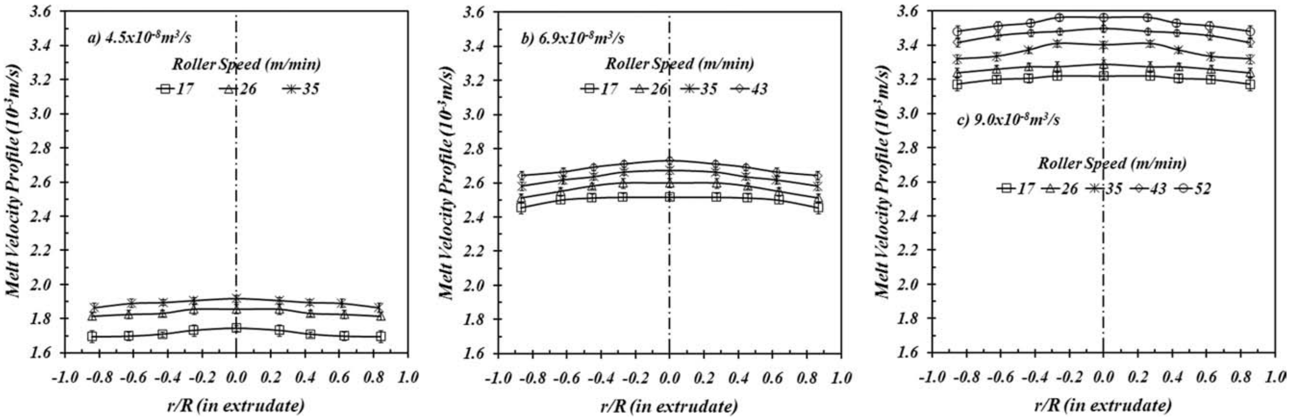

Figure 10 Melt velocity profiles for the LDPE filament from a circular die under isothermal filament stretching with increasing step-ladder roller speeds for three different volumetric flow rates: (a) 4.5 × 10^{-8}, (b) 6.9 × 10^{-8}, and (c) 9.0 × 10^{-8} m^3/s.

systems. First, the roller speeds to failure for any given volumetric flow rate between the nonisothermal and isothermal stretching conditions were different. The roller speed to failure for the isothermal stretching condition was higher than that for the nonisothermal one; this was probably related to the lower melt viscosity of the isothermal filament, which facilitated the molecular disentanglement during the flow. This could be supported by lower draw-down forces during the isothermal stretching condition, as noted by Figures 4 and 5. Second, for any given roller speed (e.g., 17–35 m/min for the volumetric flow rate of 9.0 × 10^{-8} m^3/s for the circular die and 2.7 × 10^{-7} m^3/s for the slit die), the velocity profiles for the nonisothermal filament were sharper, with the differences in the melt velocities at different $r/R/w/W$ positions being more obvious. This difference was caused by the cooling effect under the nonisothermal condition, as already discussed.

Having taken all the conditions into account, we observed that the melt velocity profiles from the slit die were greater than those from the circular die because of the higher volumetric flow rate from the extruder. It should be noted that direct comparisons of the velocity profiles obtained by these two dies for any given flow rate were not intended to be made because the volumetric flow rates obtained from these two dies were different. The reasons for using different volumetric flow rates were already mentioned in the Experimental section. One interesting point to mention was that during the experiment, we observed that for any given die geometry, volumetric flow rate, or roller speed, the melt extruded in the isothermal condition had greater swelling than that in the nonisothermal condition. The greater swelling ratio of the melt in the isothermal condition indicated some transverse flow direction of the melt, and this would have caused a lower velocity of the melt in the axial flow direction (comparing Fig. 8 with Fig. 10 and Fig. 9 with Fig. 11). This may have been one of the main reasons why the melt velocities for both die geometries under the

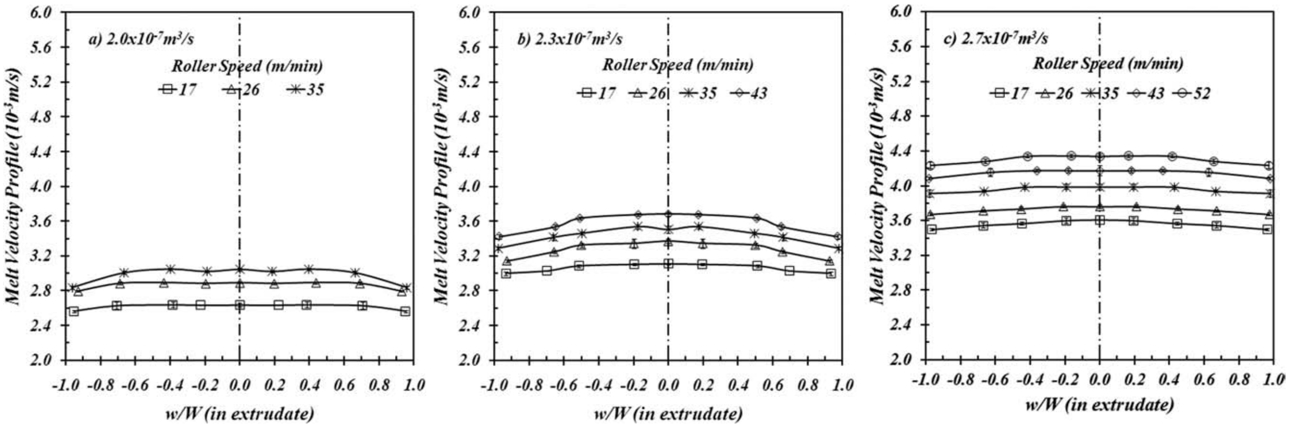

Figure 11 Melt velocity profiles for the LDPE filament from a slit die under isothermal filament stretching with increasing step-ladder roller speeds for three different volumetric flow rates: (a) 2.0 × 10^{-7}, (b) 2.3 × 10^{-7}, and (c) 2.7 × 10^{-7} m^3/s.

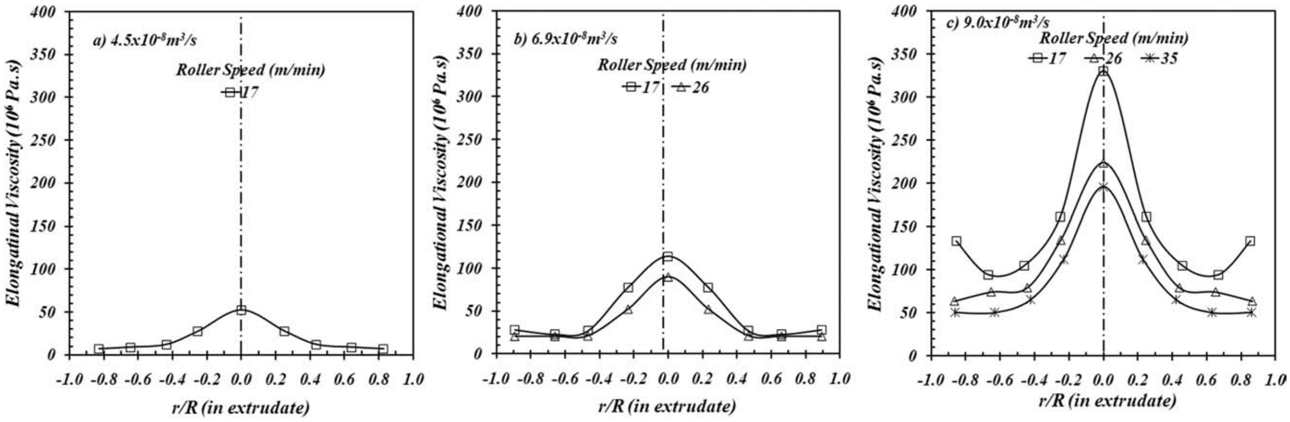

Figure 12 Elongational viscosity profiles for the LDPE filament flowing from a circular die under nonisothermal stretching for three different volumetric flow rates: (a) 4.5×10^{-8}, (b) 6.9×10^{-8}, and (c) 9.0×10^{-8} m^3/s.

isothermal condition were lower than those in the nonisothermal condition. At this point, we concluded that the melt velocity profiles across the cross section of the molten filament extruded through the circular and slit dies were affected by the temperature profiles of the filament, which were referred to as the nonisothermal and isothermal conditions during filament stretching. More obvious differences in the melt velocity profiles across the molten filament were observed for the nonisothermal filament stretching condition. If this was the case, one would expect, on the basis of eqs. (3) and (4), to have different elongational viscosities across the cross section of the molten filament.

Local elongational viscosity profiles

Nonisothermal stretching condition

In this section, we discuss the local elongational viscosity profiles across the LDPE filament diameter, which were derived and calculated, on the basis of eqs. (3) and (4), from the draw-down forces and velocity profiles with filament stretching conditions; this are given and discussed in Figures 4 and 5 and 8–11. The elongational viscosity profiles as a function of the r/R position of the LDPE filament for the circular die under the nonisothermal filament stretching condition with three different volumetric flow rates are given in Figure 12. It was found that the elongational viscosities of the molten LDPE were not uniform across the cross section of the filament diameter. The elongational viscosity appeared to decrease with increasing r/R position; it was high at the center position of the filament diameter and low near the edge of the LDPE filament. These uneven viscosities across the LDPE filament cross section seemed to be more pronounced for low roller speeds and high volumetric flow rates [see Fig. 12(c)]; this occurred because of the increased melt velocities at the filament center with increasing volumetric flow rate and roller speed, as noted in Figure 8. Another reason for the decreases in the melt viscosities of the melt as the roller speed increased was the shear heating effect,[28,29] although the melt experienced and was dominated by elongational deformation.[8,9]

An opposite result of the elongational viscosity profiles was given when the slit die was used to extrude the LDPE melt. Figure 13 shows the elongational viscosity profiles as a function of the w/W position across the cross section of the LDPE melt stretched from the slit die under the nonisothermal condition with three different volumetric flow rates. It was again observed that the elongational viscosities of the molten LDPE were not uniform across w/W of the filament; that is, the elongational viscosity profiles around the center of the melt were flat and sharply increased near the edge of the melt. The exceptionally high elongational viscosity at the filament edge was probably caused by an obvious drop in the melt velocities at this region, as shown in Figure 9. Other differences in the viscosity profiles of the LDPE filament between the circular and slit dies were the sensitivities to the volumetric flow rate and roller speed effects. It was observed that the elongational viscosity profiles of the LDPE filament from the slit die, as shown in Figure 13, were unaffected by the volumetric flow rate and roller speed effects. On the basis of the results in Figures 5 and 9, this could be reasoned by balanced or equal increases in the draw-down forces and local melt velocities with changing volumetric flow rates and roller speeds.

It would be interesting to discuss and compare the differences in the elongational viscosity profiles from the circular and slit dies in terms of the effect of temperature gradients across the molten filaments. It was clearly noted by the experimental setup that the half-width (1.5 mm) in the slit die, in which the heat transfer across the filament occurred, was relatively smaller than the radius (3 mm) of the

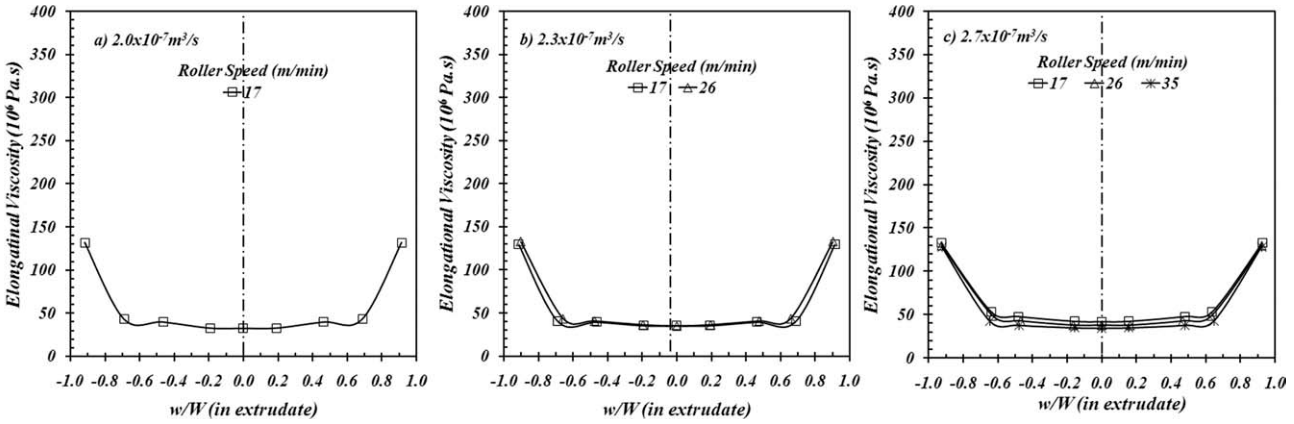

Figure 13 Elongational viscosity profiles for the LDPE filament flowing from a slit die under nonisothermal stretching for three different volumetric flow rates: (a) 2.0 × 10^{-7}, (b) 2.3 × 10^{-7}, and (c) 2.7 × 10^{-7} m^3/s.

circular die. Therefore, the heat-transfer effect, which caused the filament to cool down (warmer), in the slit die would expected to be higher. This was why the increases in the elongational viscosity around the die edge (wall) in the slit die were obvious. In the case of the circular die, the cooling effect was suppressed by the effect of the annular cross-sectional area that was used in the elongational viscosity calculations in eq. (3). That was, for a given *dr*, the area of the cross section (2π*rdr*) around the center was much lower than that near the die wall. When this was the case, on the basis of eq. (3), the elongational viscosity at the die center would increase, as observed in Figure 12.

Isothermal stretching condition

The elongational viscosity profiles as functions of the r/R position in the circular die and the w/W position in the slit die for the LDPE filament are given in Figures 14 and 15, respectively, under the isothermal stretching condition for three different volumetric flow rates. Having compared the nonisothermal condition results in Figures 12 and 13, we found that the general viscosity profiles were similar, but for any specific volumetric flow rate or roller speed, the elongational viscosities of the LDPE melt under the nonisothermal stretching condition were greater than those under the isothermal condition. This statement was found to be true and more pronounced in the circular die. For example, the elongational viscosity of the LDPE melt under the nonisothermal stretching condition (Fig. 12) at a volumetric flow rate of 9.0 × 10^{-8} m^3/s with a take-up speed of 26 m/min was 220 × 10^6 Pa s, whereas that under the isothermal stretching condition (Fig. 14) under the same volumetric flow rate and roller speed was 115 × 10^6 Pa s. However, it was observed that the differences in the elongational melt viscosities in the case of the slit die between the isothermal and nonisothermal stretching conditions were only obvious at the edge region of the LDPE filament, with the viscosity values being about 130 × 10^6 Pa s for the nonisothermal condition and around 60–80 × 10^6 Pa s for the isothermal condition. The greater elongational viscosity for the nonisothermal condition

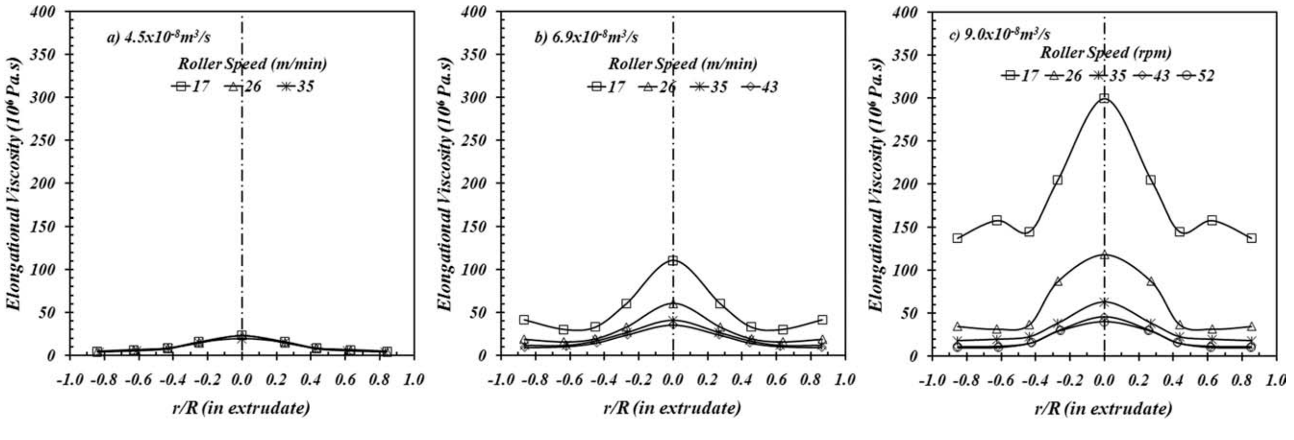

Figure 14 Elongational viscosity profiles for the LDPE filament flowing from a circular die under isothermal stretching for three different volumetric flow rates: (a) 4.5 × 10^{-8}, (b) 6.9 × 10^{-8}, and (c) 9.0 × 10^{-8} m^3/s.

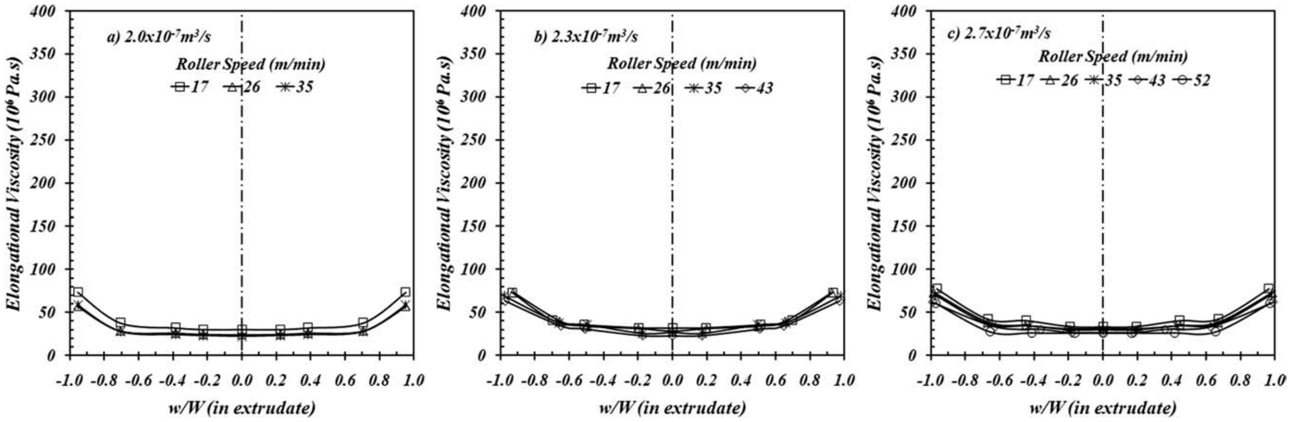

Figure 15 Elongational viscosity profiles for the LDPE filament flowing from a slit die under isothermal stretching for three different volumetric flow rates: (a) 2.0 × 10^{-7}, (b) 2.3 × 10^{-7}, and (c) 2.7 × 10^{-7} m^3/s.

corresponded very well to the findings by Muke et al.[5] One interesting aspect to note was that the elongational viscosity of the LDPE melt extruded and stretched from the circular die for any given roller speed was greater than those from the slit die. This may have been related to the higher draw-down forces and local melt velocities of the LDPE filament in the slit die, which tended to generate more shear heating effects to the melt stream and, thus, resulted in lower elongational melt viscosities in the slit die system. If this was the case, the LDPE melt in this work did not experience the strain-hardening effect during the stretching process during the experiment. This may have been because the strain rates used in this work ranged between 0.1–1.0 s^{-1}, which significantly exceeded the tension-thickening range, which has been reported to be around 10^{-3} to 10^{-1} s^{-1}.[30] Work by Padmanabhan and Macosko[31] investigated the elongational viscosity for the LDPE melt with Cogswell's analysis and suggested that the LDPE could exhibit a tension-thickening behavior only at low strain rates (ca. 10^{-1} s^{-1}).

It was also expected that for any given volumetric flow rate, the roller speed to failure for the isothermal stretching condition would be greater than that for the nonisothermal stretching condition because of the facilitating effect to the sliding molecular chains of LDPE, as mentioned earlier. This was also the case for both die design systems. The effects of the volumetric flow rate and the roller speed on the elongational viscosity profiles were found to be large with the circular die and very small for the slit die, with the latter case being related to the balanced increasing effects of the draw-down forces and melt velocities, as already discussed.

CONCLUSIONS

PCT was used successfully for simultaneous measurements of the draw-down forces (melt strength), local velocity profiles, and elongational viscosity profiles across of molten LDPE filament from either a circular or slit die under nonisothermal and isothermal conditions. The following experimental results were noted:

- The draw-down forces from both dies sharply increased at the beginning of the roller speed and then leveled off for further increasing roller speeds until the LDPE molten filament failed. The higher the volumetric flow rate was, the greater was the draw-down force that was required. For any given volumetric flow rate or roller speed, the draw-down forces in the circular die were lower than that in the slit die, and the draw-down forces under the nonisothermal condition were greater than those for the isothermal condition.
- The velocity profiles of the melt were equalized at the die exit under the no-stretching condition but were not uniform when the melt was stretched. The melt velocities at any local point across the filament cross section appeared to increase with increasing volumetric flow rate and roller speed. The overall melt velocities in the slit die and under the nonisothermal condition were greater than those in the circular die and under the isothermal condition.
- The elongational viscosity profiles of the LDPE were not uniform across the cross section of the filament; this was pronounced when the circular die, low roller speeds, and high volumetric flow rates were used. The elongational viscosities of the LDPE melt extruded and stretched from the circular die ad under the nonisothermal condition were greater than those from the slit die and under isothermal stretching condition. The effects of the volumetric flow rate and the roller speed on the elongational viscosity profiles

were large with the circular die and rather small with the slit die.

References

1. Baldi, F.; Franceschini, A.; Riccò, T. Rheol Acta 2007, 46, 965.
2. Gupta, R. K.; Bhattacharya, S. N. J Polym Eng 2007, 27, 89.
3. Yamaguchi, M.; Suzaki, K.-I. J Appl Polym Sci 2002, 86, 79.
4. Wagner, M. H.; Bastian, H.; Bernnat, A.; Kurzbeck, S.; Chai, C. K. Rheol Acta 2002, 41, 316.
5. Muke, S.; Ivanov, I.; Kao, N.; Bhattacharya, S. N. J Non-Newt Fluid Mech 2001, 101, 77.
6. Baldi, F.; Franceschini, A.; Bignotti, F.; Tieghi, G.; Riccò, T. Rheol Acta 2009, 48, 73.
7. de Jeśus, T.; Guadarrama-Medina, T. J.; Pérez-González, J.; de Vargas, L. Rheol Acta 2005, 44, 278.
8. Sitticharoen, W.; Intawong, N.-T.; Sombatsompop, N. Polym Polym Compos 2010, 18, 359.
9. Harnnarongchai, W.; Intawong, N.-T.; Sombatsompop, N. J Macromol Sci Phys 2011, 50, 1074.
10. Harnnarongchai, W.; Sitticharoen, W.; Intawong, N.-T.; Sombatsompop, N. J Vinyl Addit Technol 2011, 17, 164.
11. Wei, X.; Collier, J. R.; Petrovan, S. J Appl Polym Sci 2007, 104, 1184.
12. Wagner, M. H.; Kheirandish, S.; Yamaguchi, M. Rheol Acta 2004, 44, 198.
13. Pérez, R.; Rojo, E.; Fernández, M.; Leal, V.; Lafuente, P.; Santamaría, A. Polymer 2005, 46, 8045.
14. Robledo, N.; Vega, J. F.; Nieto, J.; Martínez-Salazar, J. J Appl Polym Sci 2011, 119, 3217.
15. Wagner, M. H.; Kheirandish, S.; Stange, J.; Münstedt, H. Rheol Acta 2006, 46, 211.
16. Kozlowski, M.; Szczurek, S.; Szczurek, T.; Frackowiak, S. Int J Mater Forum 2008, 1, 751.
17. Li, T. Q.; Wolcott, M. P. Compos A 2004, 35, 303.
18. Dintcheva, N. T.; Arrigo, R.; Morreale, M.; Mantia, F. P. L.; Matassa, R.; Caponetti, E. Polym Adv Technol 2010. DOI: 10.1002/pat.1648.
19. Intawong, N.-T.; Sombatsompop, N. Polym Eng Sci 2004, 44, 1960.
20. Schneider, C.; Schwetz, M.; Münstedt, H.; Kaschta, J. Mech Time-Dependent Mat 2004, 8, 215.
21. Hertel, D.; Valette, R.; Münstedt, H. J Non-Newt Fluid Mech 2008, 153, 82.
22. Goutille, Y.; Guillet, J. J Non-Newt Fluid Mech 2002, 102, 19.
23. Schuberth, S.; Münstedt, H. Rheol Acta 2008, 47, 111.
24. Kim, H.; Chung, K.; Youn, J. R. Fiber Polym 2000, 1, 37.
25. Intawong, N.-T.; Sombatsompop, N. Polym Eng Sci 2004, 44, 2298.
26. Sombatsompop, N.; O-Charoen, N. Polym Adv Technol 2003, 14, 699.
27. Meerveld, J. V.; Hütter, M.; Peters, G. W. M. J Non-Newt Fluid Mech 2008, 150, 177.
28. Tan, L. P.; Joshi, S. C.; Yue, C. Y.; Lam, Y. C.; Hu, X.; Tam, K. C. Acta Mater 2003, 51, 6269.
29. Wapperom, P.; Hassager, O. Polym Eng Sci 1999, 39, 2007.
30. Feigl, K.; Tanner, F. X.; Edwards, B. J.; Collier, J. R. J Non--Newt Fluid Mech 2003, 115, 191.
31. Padmanabhan, M.; Macosko, C. W. Rheol Acta 1997, 36, 144.