Radial Velocity Profiles and Melt Strength of LDPE Melt under Elongational Flow in Circular Die

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Abstract

An experimental arrangement to simultaneously measure melt strength, velocity profiles, and elongational viscosity profiles across the filament diameter for low-density polyethylene (LDPE) under non-isothermal and isothermal conditions was proposed. The proposed experimental rig was based on parallel co-extrusion technique (PCT) of colored LDPE melt-layers into uncolored melt flowing from the barrel into and out a circular die forming a continuous filament before being pulled down by mechanical rollers until the filament failed. The results suggested that the higher volumetric flow of the LDPE melt the greater the drawdown force required to filament failure, and thus, higher local melt velocities and elongational viscosities. The isothermal filament stretching gave lower drawdown forces with greater roller speed to failure than the non-isothermal filament stretching. Without stretching the LDPE filament, the melt velocities across the filament diameter were similar, forming plug-like velocity profiles. But, the differences in melt velocities across the filament diameter became apparent under the non-isothermal filament stretching condition. This behavior was also the case for elongational viscosity profiles. The velocity and elongational melt viscosity profiles were also affected when being extruded at high volumetric flow rates and being stretched at high roller speeds used.

1. Introduction

Melt spinning is a common process to produce melt filament and is also used for measurements of melt properties under elongational flow and deformation. The melt properties in this process are referred to as melt strength, elongational stress and strain and elongational viscosity. Baldi et al. [1] found that the melt strength for high-density polyethylene under melt spinning experiment increased with increasing extrusion rate. Sitticharoen et al. [2] found that the drawdown force for low-density polyethylene melt increased with increasing roller speeds, the take-up style and the higher the volumetric flow rate the greater the drawdown force. Gupta and Bhattacharya [3] investigated the effects of die geometries and extrusion rates on melt strength of polypropylene (PP) and high-melt strength PP using the Rheotens. The results indicated that die diameter had greater effect on melt strength than the die length and the higher the extrusion rate the greater the melt strength. Many researchers [1-7] have discovered methods and techniques to investigate the velocity profiles and flow patterns of flowing polymer melt both in shear and elongational flows while the wildly used technique is called "Laser-Doppler velocimetry (LDV)". This technique is relatively accurate, but very expensive and complicated to use [4-5]. Intawong and Sombatsompop [6] studied the radial velocity profiles of polystyrene melt in shear flow using a parallel co-extrusion technique (PCT), and found that the radial velocity profiles of the melt changed continuously with extrusion time. In elongational flow, the melt velocities along spin-line were found to increase with increasing roller speed take-up velocity [7].

Based on the work by Intawong and Sombatsompop [6], it was suggested that the velocities of the melt across the flow channel were different although they would equalize as being exited the die. However, during the free flow, the melt at the filament skin may encounter a cooling effect. This would probably make the velocity gradient across the filament diameter. If this was the case, the elongational strain would be un-even elongational strain and elongational viscosity across the filament diameter. But, these kinds of information have not yet been fully discussed and understood. Therefore, this work was proposed to simultaneously measure melt strength, velocity profiles, and elongational viscosity profiles across the filament diameter for low-density polyethylene (LDPE) under non-isothermal and isothermal conditions. The study was carried out using an experimental rig which was specially designed and developed based on a parallel coextrusion technique (PCT) [6]. The filament diameter for LDPE was evaluated the drawdown forces (mechanical strength), the radial velocity profiles across the filament diameter and radial elongational viscosity by the effects of volumetric flow rates and roller speed.

2. Experimental

2.1 Raw materials

1. Low-density polyethylene (LD1905FA), with a melt flow rate of 5 g/10 min was used a skin layer, and supplied by Thai Polyethylene Co., Ltd. (Bangkok, Thailand). Red masterbatch (3.0%) was used as multi-core layers, supplied by Clariant Co., Ltd., (Bangkok, Thailand), in co-extrusion process.
2. Corn particles (0.1%) were relatively small of 210 μm, the maximum moisture content allowed being less than 5%, and used a core layer.

2.2 Design and manufacture of the experimental apparatus

An experimental technique in Figure 1 was proposed to simultaneously measure melt strength (as drawdown force), velocity profiles, and elongational viscosity profiles across the filament diameter for LDPE under non-isothermal and isothermal conditions. The non-isothermal was referred to as the filament being cooled down by ambient air whereas the isothermal was referred to as the temperature of the filament being 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 was two infrared heaters (500 W, 220 V) and a DD6 temperature controller system was used. The temperatures within the heating chamber were exactly the same. The velocity profiles across the filament diameter were visualized by a color video-camera for further calculations of the radial elongational viscosity across the filament diameter for LDPE.

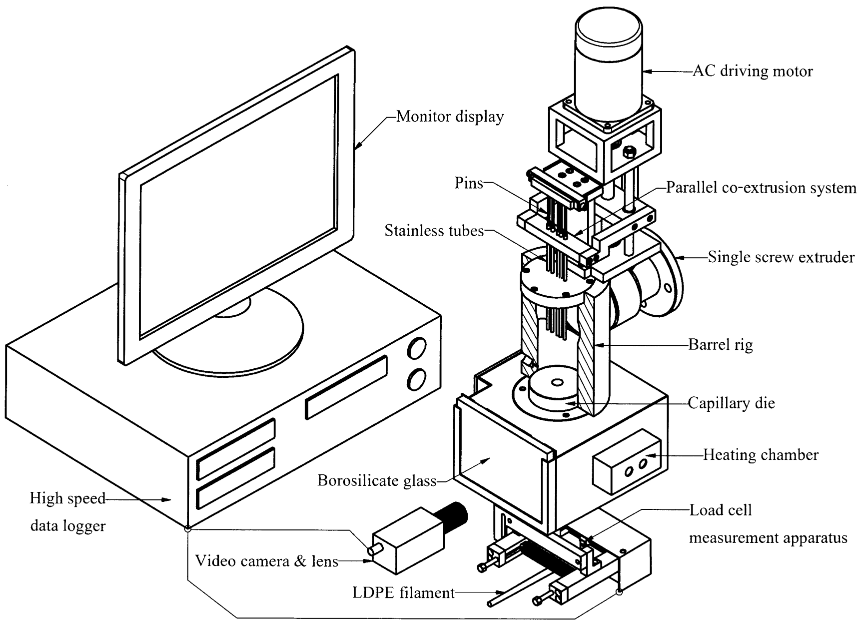


Figure 1. Schematic of radial velocity profiles and melt strength measurement

2.3 Measurements of elongational viscosity and velocity profiles

The velocity profile measurements were carried out and monitored by recording the times taken for the natural corn particles which inserted into and flow along the melt layers to travel for a given distance in the LDPE filament. All the experimental data were recorded and displayed using a high speed data-logging and recording system and a personal computer. The reported experimental data were averaged from at least five independent determinations. The average melt velocity of each colored layer at the reduced radial (r/R) position across the filament diameter (v_n) could be calculated using Eq. 1 where L_0 is the observed length of the measurement ($15x10^{-3}$m) and t_c is the time.

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

Thus, elongational viscosity (λ) is expressed by Eq. 2 [1] where F is the average drawdown force by increasing step-ladder roller speeds, L is the length of spin-line, Q is the volumetric flow rate from the screw extruder, and ε_E is elongational strain which is defined as $\ln\left(\frac{v_n}{v_0}\right)$, v_0 is the melt velocity at die exit. The viscosities for LDPE melt were calculated using the melt velocities (v_n) at any radial points across the filament diameters.

$$\lambda = \frac{F.L}{Q \cdot \varepsilon_E} \tag{2}$$

2.4 Test variables

In this work, the test variables were volumetric flow rate and roller speed. The volumetric flow rate was varied from 4.5 x 10^{-8} to 9.0 x 10^{-8} m^3/s. The length of the spin-line was fixed at 310 mm. The die temperature used was 150 ^oC.

3. Results and Discussion

3.1 Effect of corn particles and LDPE masterbatch on melt flow properties

Since this work used corn particles as foreign object to follow the velocity of the LDPE melt and LDPE masterbatch for co-extrusion it was essential to determine whether the corn particles and the LDPE masterbatch affected the flow properties of the LDPE melt. Figure 2 shows the results of apparent wall shear stress and apparent wall shear rate for LDPE melt with and without additions of corn particles and LDPE masterbatch at 150°C. It was found that the flow curves for both LDPE systems were very similar, the differences being within the experimental error of ± 4.9%. It was therefore confirmed that the corn particles and the masterbatch did not affect the flow properties of the molten LDPE.

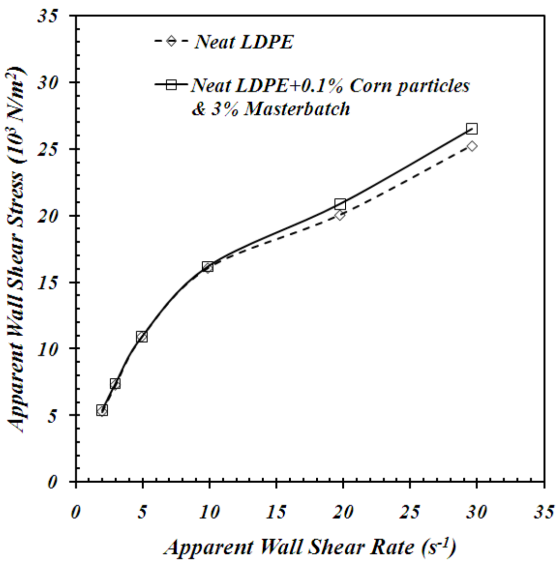


Figure 2. Flow curve of molten LDPE with and without the addition of corn particles and LDPE masterbatch at a die temperature of 150°C

3.2 Mechanical properties of LDPE extrudate filament

Figure 3 shows the drawdown force against extrusion time with increasing step-ladder roller speeds for LDPE extrudate filament under non-isothermal and isothermal conditions for three different volumetric flow rates. The results indicated that the drawdown force sharply increased at the very beginning of the roller speed take-up and then stabilized at further increasing roller speeds until the LDPE extrudate filament had failed. The changes in drawdown forces with roller speed effect could be explained by molecular entanglements and long chain branching of LDPE as detailed in our previous work[2]. For the effect of volumetric flow rate from the extruder, it was found that the higher the volumetric flow rate the greater the drawdown force required. This claim was supported by the works of Gupta and Bhattacharya [3]. The extrudate filament with higher volumetric flow rate tended to generate greater stored energies in the molten polymer and this was caused enhanced the drawdown force. This view could be supported by Baldi et al [1]. Comparing the results between Figures 3a and 3b, it was noticeable that for any given roller speeds, the drawdown forces for the extrudate filament under non-isothermal condition were greater than those for the isothermal condition. This was caused by the fact that the extrudate filament under non-isothermal condition tended to solidify during filament stretching, especially on the surface of the extrudate filament, and this then led to significantly increased filament viscosity and thus increased drawdown forces.

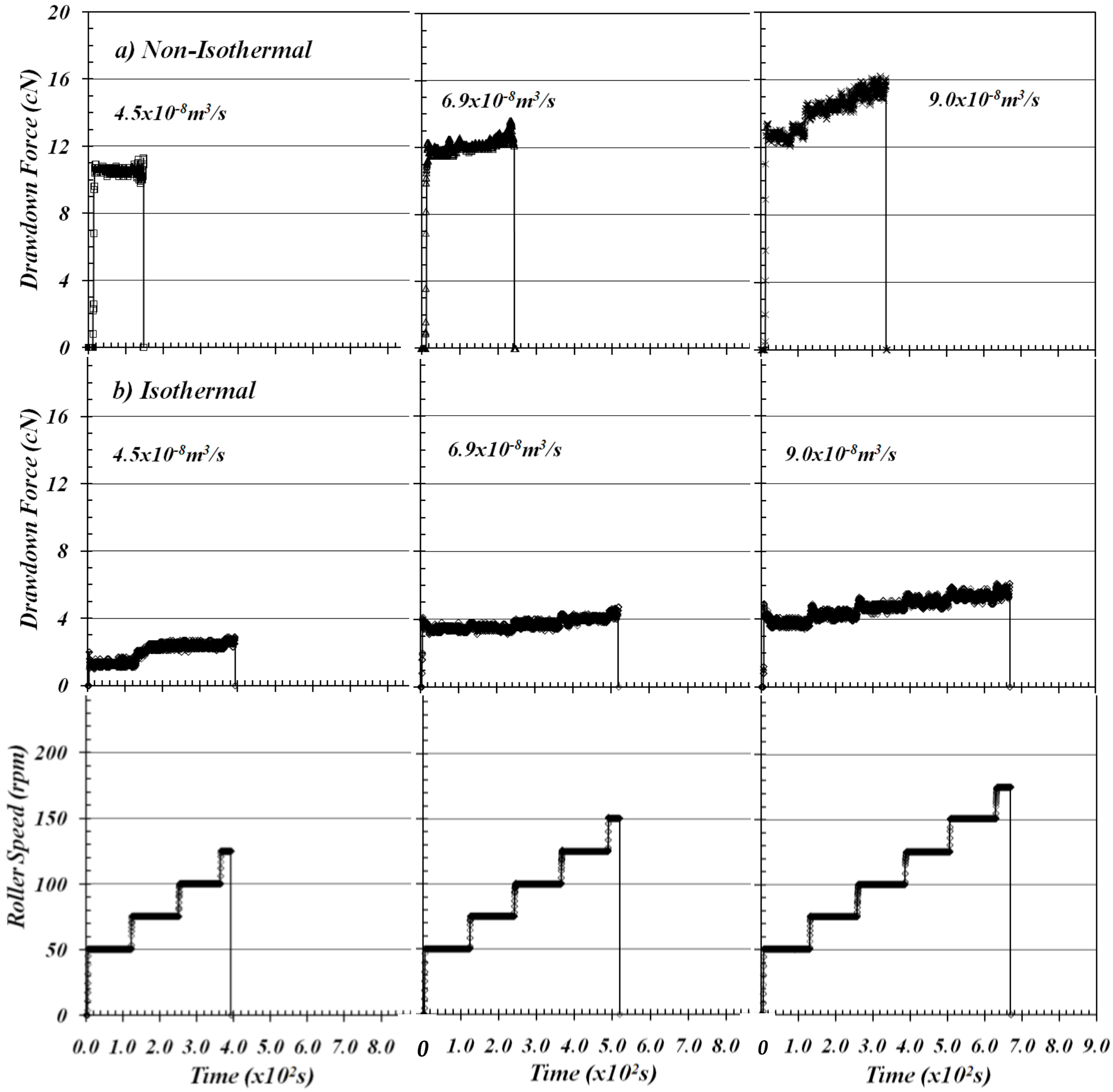


Figure 3. Drawdown force VS time as a function of roller speed for LDPE extrudate filament for three different volumetric flow rates. (a) non-isothermal filament stretching (b) isothermal filament stretching

However, it was found that the extensional strains (or roller speed to failure) for the extrudate filament under isothermal condition was greater than those under non-isothermal condition. This may be expected since the melt viscosity of the extrudate filament under isothermal condition was lower because the molecules were not restricted by the cooling effect [2,6].

3.3 Radial velocity profile of LDPE filament

Figure 4 shows the radial velocity profiles as a function of reduced radial (r/R) position across the diameter for LDPE extrudate filament under non-isothermal and isothermal conditions without stretching by the rollers for three difference volumetric flow rates. It was found that the velocity profiles of the extrudate filament for both cases were plug-like, the melt velocities across the filament diameters being very similar. This was expected as the melt existed the die, the explanations being found elsewhere [6]. The greater the volumetric flow rate from the extruder the higher the melt velocities across the filament diameter. It was also observed that for a given volumetric flow rate, the melt velocities for the extrudate filament under isothermal condition were higher than those under non-isothermal condition. This was because the melt viscosity of the isothermal filament was lower.

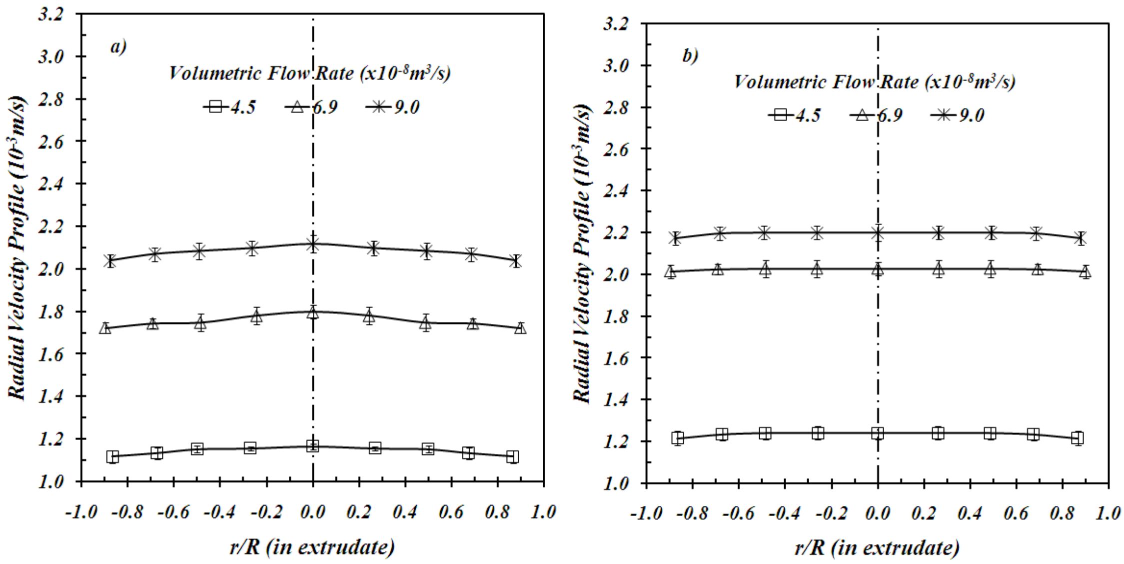


Figure 4. Radial velocity profiles for LDPE filament *without* filament stretching for three difference volumetric flow rates. (a) non-isothermal condition (b) isothermal condition

Figure 5 shows the radial velocity profiles as a function of reduced radial (r/R) position across the diameter for LDPE extrudate filament under non-isothermal 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 decreased with increasing r/R position, high melt velocity at the centre position of the filament diameter and low near the edge of the filament strand. As the roller speed increased the melt velocities at any radial points across the filament diameter increased. This view could be supported by the work of Meerveld et al. [7] who stated that the velocity profiles along spin-line increased with increasing the roller speed take-up velocity. The radial melt velocities also increased with increasing volumetric flow rate. It was interesting to note that the roller speed to failure increased with increasing volumetric flow rate, i.e., the filament at volumetric flow rate of 4.5×10^{-8} m^3/s failed at the roller speed of

50 rpm whereas that at volumetric flow rate of $9.0\text{x}10^{-8}$ m^3/s failed at the roller speed of 100 rpm. This was associated with the stored energies within the filaments during the extrusions.

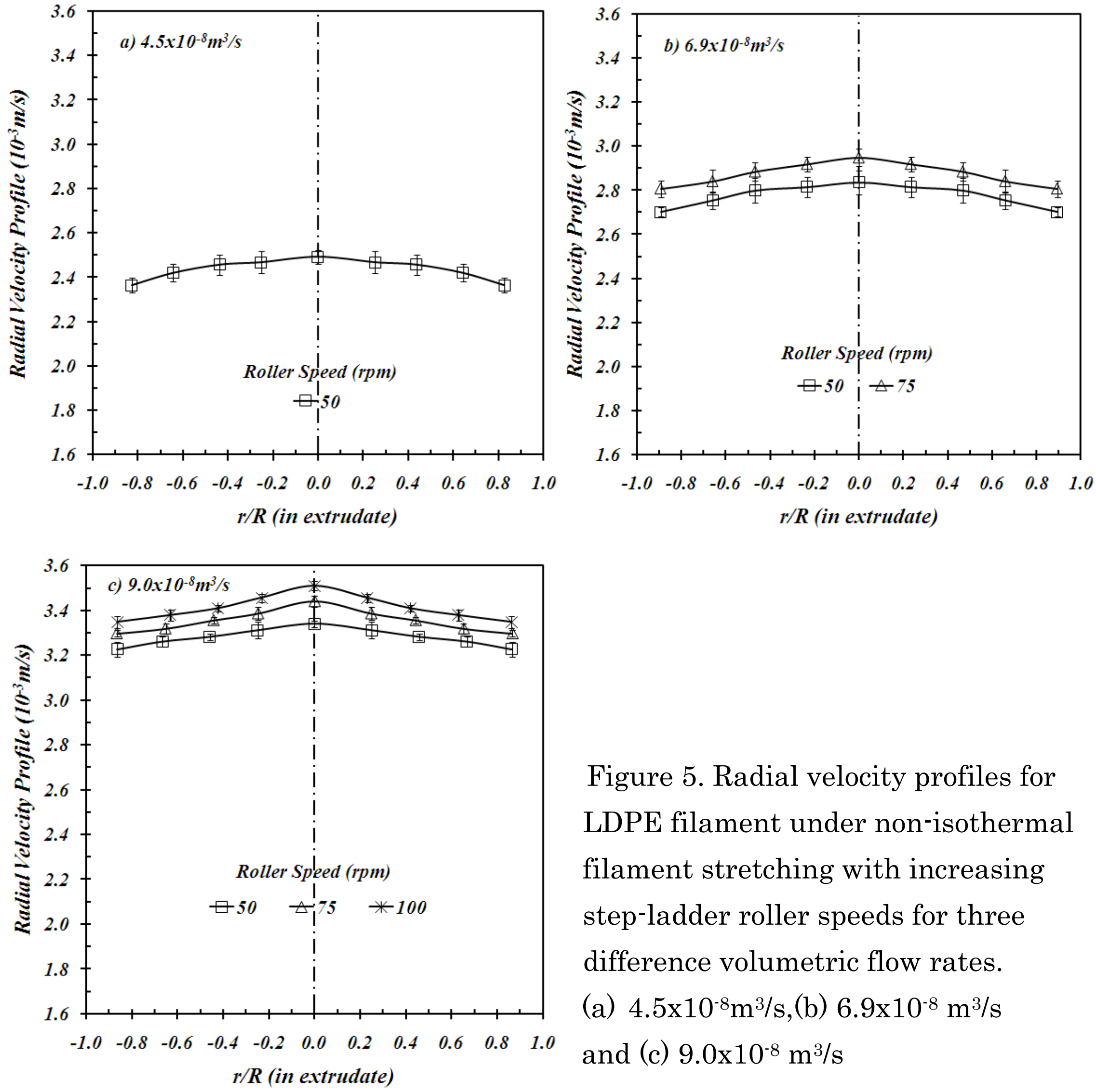
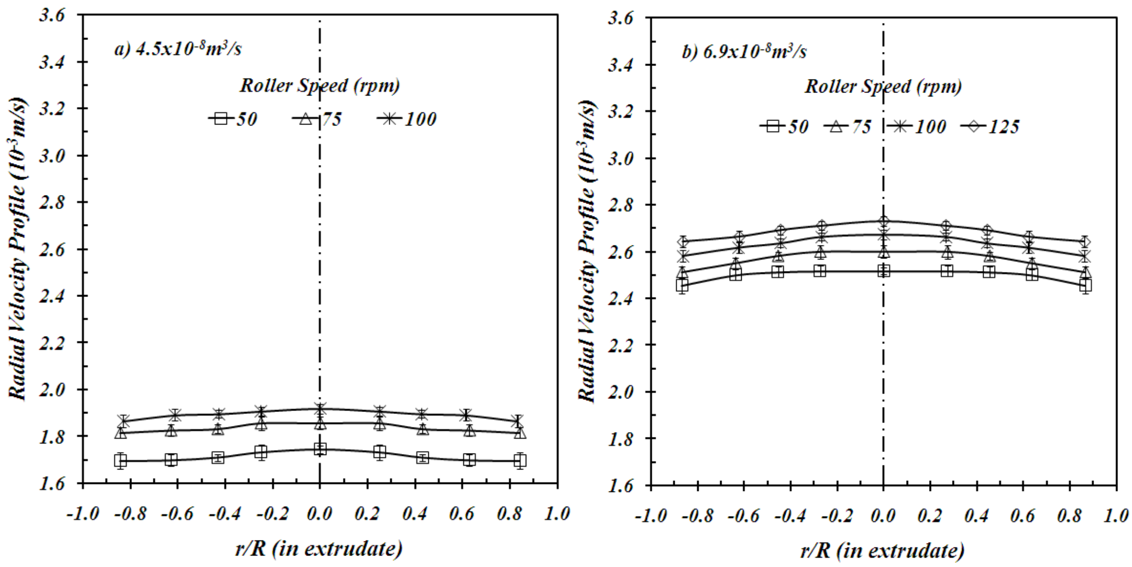


Figure 5. Radial velocity profiles for LDPE filament under non-isothermal filament stretching with increasing step-ladder roller speeds for three difference volumetric flow rates. (a) $4.5\text{x}10^{-8}m^3/s$,(b) $6.9\text{x}10^{-8}$ m^3/s and (c) $9.0\text{x}10^{-8}$ m^3/s

Figure 6 shows the radial velocity profiles for LDPE extrudate filament under isothermal filament stretching condition with increasing step-ladder roller speeds using three difference volumetric flow rates. The general patterns of the melt velocity profiles as a function of reduced radial position, volumetric flow rate and roller speed were very similar to those in the non-isothermal stretching in Figure 5. However, two differences were noted. First, the roller speeds to failure for any given volumetric flow rates between non-isothermal and isothermal stretching conditions were different. The roller speed to failure for isothermal faliment stretching condition was higher than that for non-isothermal one. This was probably related to the fact that the melt viscosity of the isothermal filament was lower and facilitate the molecular disentanglement during the flow. But, noted by Figure 3, the drawdown forces during the stretching for the isothermal condition were much lower. Second, for any given roller speeds (such as 50-100 rpm for the volumetric flow rate of 9.0×10^{-8} m^3/s) the velocity profiles for the non-isothermal filament were sharper, the differences in melt velocities at different radial positions being more obvious. This difference was caused by the cooling effect under the non-isothermal condition. One interesting point to mention was that for any given volumetric flow rates and roller speeds, the radial velocities of the melt in isothermal condition were lower than those in non-isothermal condition. This was because, the melt extruded in the isothermal condition was observed to greater swelling than that in the non-isothermal.



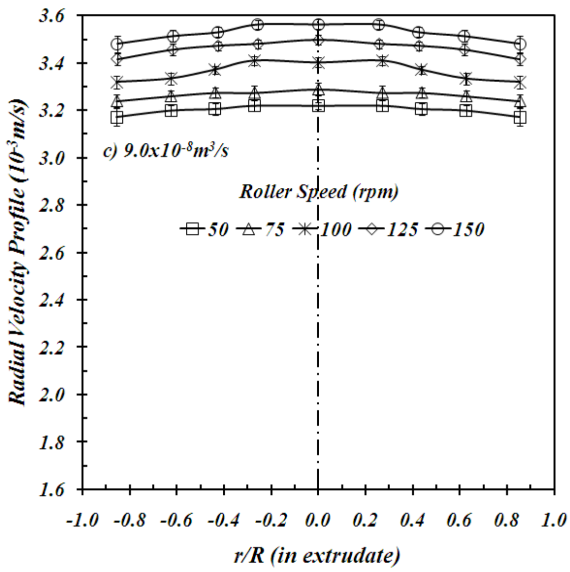


Figure 6. Radial velocity profiles for LDPE filament under isothermal filament stretching with increasing step-ladder roller speeds at 150 °C for three difference volumetric flow rates. (a)4.5×10^{-8} m^3/s, (b) 6.9×10^{-8} m^3/s and (c) 9.0×10^{-8} m^3/s

In summary, taking account the experimental results in Figures 5 and 6, it was suggested that the melt velocity profiles across the filament diameter were affected by the temperature profiles of the filament which were referred to as non-isothermal and isothermal condition during filament stretching. The melt velocities of the extrudate were different across the filament diameter, the differences being more pronounced in the case of non-isothermal filament stretching. If this was the case, one would expect to have different radial extensional (elongational) viscosities across the filament diameter.

3.4 Radial elongational viscosity of LDPE extrudate filament

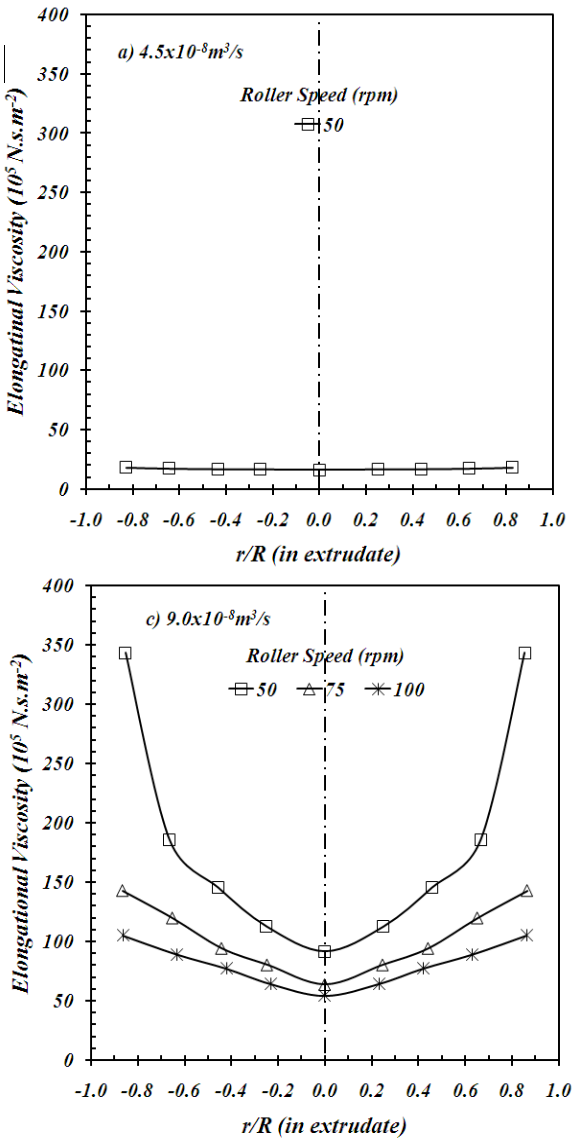


Figure 7. Radial elongational viscosity profiles for LDPE filament under non-isothermal stretching for three difference volumetric flow rates. (a) 4.5×10^{-8} m^3/s, (b) 6.9×10^{-8} m^3/s and (c) 9.0×10^{-8} m^3/s

This section determined the radial elongational viscosity profiles across LDPE filament diameter. Figures 7 and 8 show radial elongational viscosity profiles as a function of reduced radial (r/R) position across the diameter for LDPE filament under non-isothermal and isothermal condition, respectively, at three difference volumetric flow rates. In general, it was found that the elongational viscosity profiles were flat when low volumetric flow rate ($4.5x10^{-8}$ m^3/s and $6.9x10^{-8}$ m^3/s) or low roller speeds (50-75 rpm). The differences in elongational melt viscosities became more obvious high volumetric flow rate with low roller speeds (Fig. 7c) or low volumetric flow rate with high roller speeds (Fig. 8b). Considering the same volumetric flow rate ($9.0x10^{-8}$ m^3/s) at the same range of roller speed (50-100 rpm) for non-isothermal (Fig. 7c) and isothermal condition (Fig. 8c), it was found that the differences in melt viscosities for the non-isothermal condition were greater and more pronounced. High melt viscosities near the filament edge (high r/R positions) were caused by to the cooling effect in the non-isothermal filament stretching.

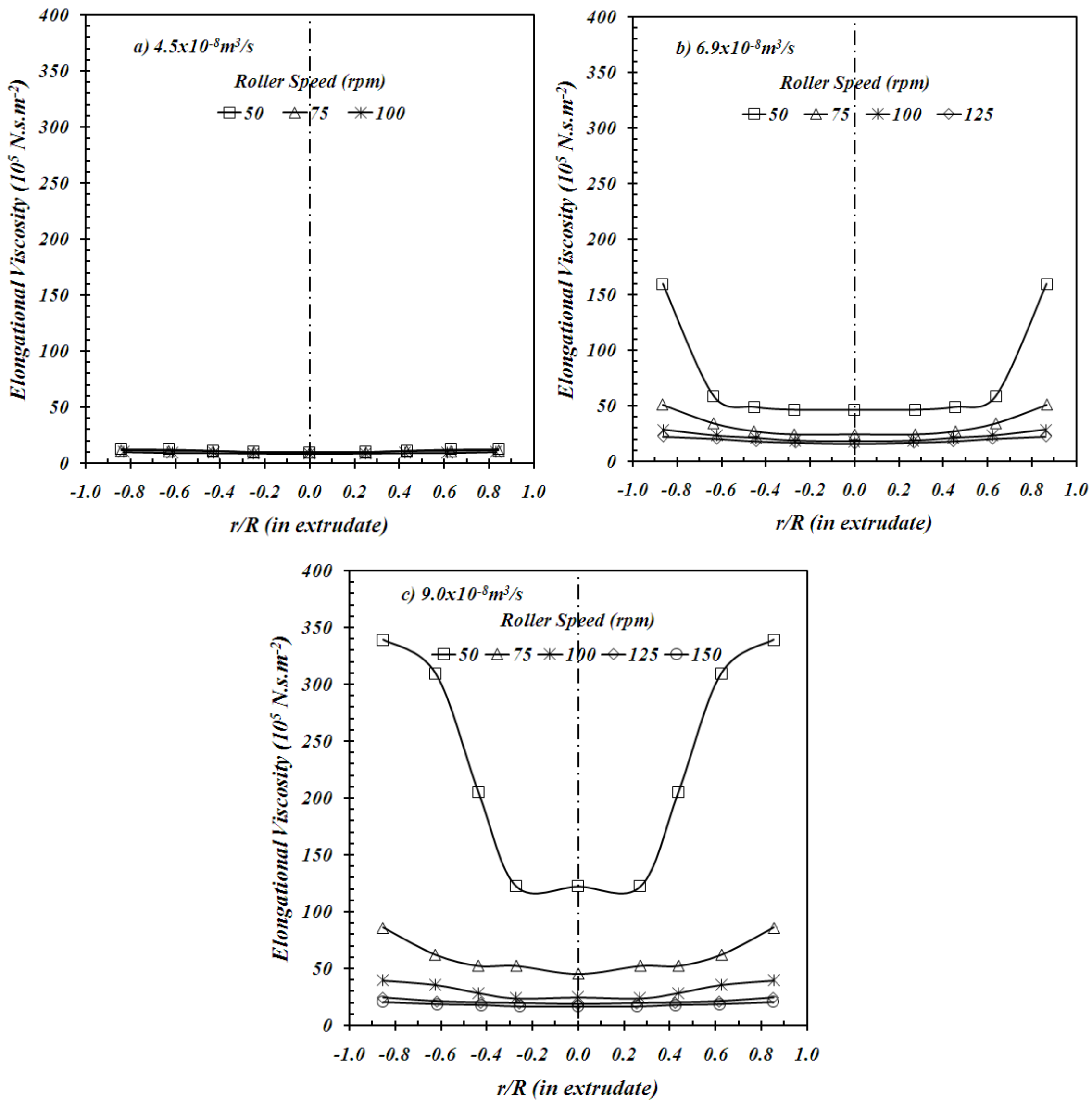


Figure 8. Radial elongational viscosity profiles for LDPE filament under isothermal stretching for three difference volumetric flow rates. (a) $4.5x10^{-8}$ m^3/s, (b) $6.9x10^{-8}$ m^3/s, and (c) $9.0x10^{-8}$ m^3/s

4. Conclusions

The parallel co-extrusion technique (PCT) could be used for simultaneous measurements of melt strength, radial velocity profiles and radial elongational viscosity profiles across the filament diameter for LDPE under non-isothermal and isothermal conditions. The results suggested that the higher the volumetric flow rate the greater the drawdown force required to filament failure, and higher local melt velocities and elongational viscosities. Greater drawdown forces with lower roller speed to failure were observed for the LDPE filament under non-isothermal condition as compared with those under isothermal condition. Without stretching the LDPE filament, the velocity profiles of the LDPE melt were plug-like. However, the melt velocities across the filament diameter were different, the effect being more obvious for non-isothermal stretching condition. The volumetric flow rate and roller speed had a significant effect on velocity and elongational viscosity profiles.

Acknowledgments

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