Study of Instabilities in Film Blowing

A. Ghaneh-Fard, P. J. Carreau, and P. G. Lafleur
Centre de Recherche Appliquée sur les Polymères (CRASP), Dept. of Chemical Engineering, Ecole Polytechnique, Montreal, Quebec, H3C 3A7 Canada

Bubble instabilities observed in film blowing using four different polyolefins are discussed: high-density polyethylene (HDPE), low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE), and polypropylene (PP). Special attention is given to the effect of the frost line height on the bubble stability, an effect mostly ignored in the literature. A video-camera system was used to record the bubble shape and oscillations. In general, three forms of instabilities and combinations were observed: (1) axisymmetric periodic variations in the bubble diameter; (2) helical motions of the bubble; and (3) variations in the position of the solidification line. The four resins show different stability behaviors. The LDPE has the most stable operating space and the PP is the most unstable one. No correlation was observed between bubble stability and oscillatory shear rheological properties of the resins. Instability is enhanced by increasing take-up ratio, increasing blow-up ratio, and decreasing frost line height. Furthermore, for the LDPE, some operating points were not attainable and multiple steady states were observed. Our results are in a poor agreement with the predictions Cain and Denn's 1988 analysis.

Introduction

Blown film extrusion, sketched in Figure 1, is an important polymer processing operation and is used to produce most of the plastic films. The molten polymer is extruded at a constant flow rate through an annular die. The film is deformed axially by the tension of the take-up device and circumferentially by the introduction of air inside the polymer tube.

A stable bubble is a requirement not only for the continuous operation of the process but also for the production of an acceptable film (Fleissner, 1988). The restriction of stable operating conditions also limits the rate of production and, due to process/physical property interactions, limits the attainable range of physical properties (Sweeney et al., 1992).

Instability in film blowing was first reported by Ast (1974) and Han and Park (1975). Han and coworkers (Han and Park, 1975; Han and Shetty, 1977) observed that lowering the extrusion temperature stabilized the blown bubble for high-density polyethylene (HDPE) and low-density polyethylene (LDPE). The work of White and coworkers (Kanai and White, 1984; Minoshima and White, 1986; White and Yamane, 1987) is the most extensive. Comparing bubble stability of different polyethylene (PE) resins, Minoshima and White (1986) have concluded that the long-chain branched PEs are the most stable, followed by the broad-distribution linear PEs. The narrower molecular weight distribution PEs are the most unstable. In other words, LDPE has the widest stable operating region and it has been attributed to the strain-hardening behavior of LDPE in elongational flow. Minoshima and White (1986) have also examined the influence of molecular structure of PE on draw resonance in melt spinning, which is kinematically similar to film blowing. LDPE was again the most stable, but broadening molecular weight distribution in linear PEs was destabilizing for melt spinning. Ghijsels et al. (1990) have suggested that in film blowing a small axial take-up force is needed to stretch the low melt strength film in the absence of strain hardening, as for linear low-density polyethylene (LLDPE). In this situation a low-tension bubble becomes sensitive to surrounding air flows and gravity forces, leading to bubble instabilities. Improving stability behavior of LLDPE by using a dual-iris cooling system or by blending with LDPE has been shown by Obijeski and Pruitt (1992). Sweeney et al. (1992) have demonstrated that the video analysis system is an effective, noncontact, real-time device for quantifying instabilities during film blowing. In another study, Sweeney and Campbell (1993) have recently observed that there is a strong level of interaction between various process parameters affecting the bubble stability.

Correspondence concerning this article should be addressed to P. J. Carreau.

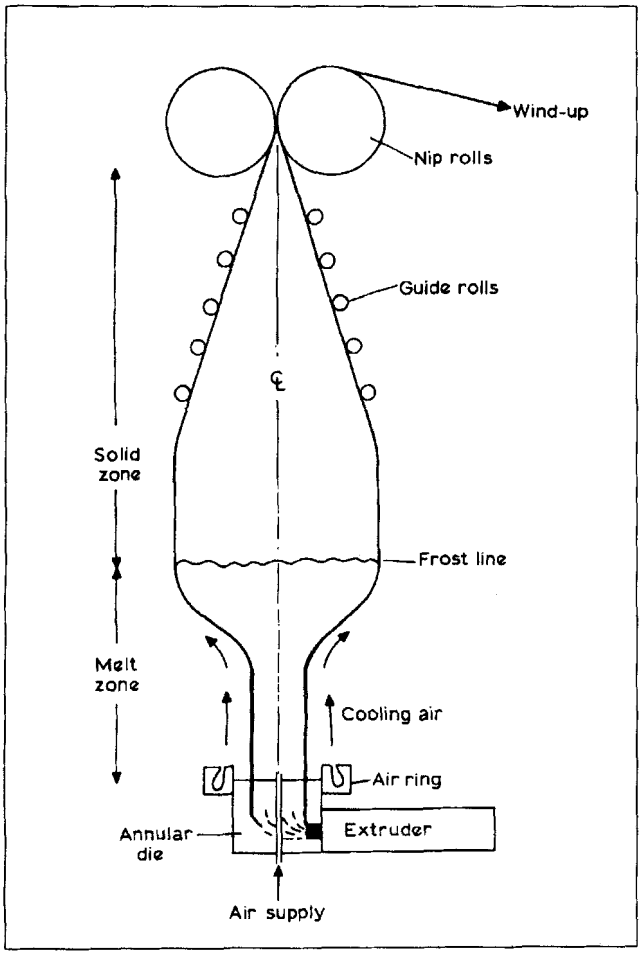

Figure 1. Blown film extrusion.

We are aware of only two theoretical studies on bubble stability, those of Yeow (1976) and Cain and Denn (1988). They both used an isothermal Newtonian model and a linear stability analysis with different numerical techniques. Cain and Denn have also carried out a mathematical analysis of stability assuming that the melt is an upper convected Maxwell fluid. However, because of these simplifying assumptions, these models do not show a good agreement with experimental observations. This will be discussed later.

The main objective of this study is to determine the stable operating space for different polyolefins. The stability behavior of three different polyethylene resins as well as of a polypropylene (PP) resin was studied using a quantitative criterion. Different forms of instability are defined and the effect of film blowing parameters are illustrated. These parameters are the frost-line height (FLH), shown in Figure 1, the blow-up ratio (BUR), defined as the ratio of the final bubble diameter to the die diameter, and the take-up ratio (TUR), which is the ratio of the take-up velocity to the extrudate velocity at the die exit. BUR and TUR are obviously two key parameters of the film blowing process. Increasing the values of these parameters is explicitly desirable in a commercial film production. On the other hand, even though FLH does not control the film geometry, its effect on final film properties cannot be denied, as it is a response to cooling conditions. That is why we have examined the influence of FLH on bubble stabilities. This article is a part of an extensive study of film blowing being carried out in our laboratory. Future work will focus on the elongational properties of polymers determined *in situ* during film blowing using a rheo-optical technique.

Experimental Studies

Materials

Four different film-grade polyolefins were used in this study: a HDPE, an LDPE, an LLDPE, and a PP. The HDPE and the LLDPE were supplied by Du Pont, the LDPE by Novacor, and the PP by Himont. Densities, melt indices (*MI*) and the available molecular characteristics of the four polymers are summarized in Table 1.

Rheological measurements

The dynamic rheological properties, storage and loss moduli, G' and G'', complex viscosity, η^*, were measured using a CSM Bohlin rheometer in a concentric disk configuration under a dry nitrogen atmosphere. The frequency used ranged from 0.001 to 30 Hz, and the applied stress was adjusted to maintain the experiments in the linear domain. Measure-

Table 1. Materials Used in This Study and Main Characteristics*

Polymer	Supplier	MI dg/min	Density kg/m^3	η_0 Pa·s	M_w kg/kmol	M_w/M_n	M_z/M_w
HDPE	Du Pont 16A	0.25	946	237,000**	140,000	7.0	10.0
LDPE	Novacor LF0222-B	2.2	922	10,000**	—	—	—
LLDPE	Du Pont 12J1	1.0	924	17,100**	100,000	5.8	3.4
PP	Himont 6631	1.8	902	3,030†	—	—	—

*Molecular characteristics of the LDPE and PP were not available; **at 180°C; and † at 240°C.

ments were carried out at a temperature of 180°C for the HDPE, the LDPE, and the LLDPE, and 240°C for the PP. Concentric disks of diameter equal to 25 mm with a gap between 1 and 1.2 mm were used for all measurements.

Blown film extrusion

A 45-mm Killion single-screw extruder with a helical blown film die (outer diameter = 50.82 mm and die gap at exit = 680 μm) was used in this study. The extrusion was carried out at a temperature of 180°C for the polyethylenes and 240°C for the PP. It was found that for our blown film system the PP was not processable at 230°C and was very unstable at 250°C. The polymer flow rate was maintained at about 6.8 kg/h for all experiments.

The film blowing process was initiated by grabbing the tube of molten polymer, leaving the die, and making it pass over the nip rolls. Inflation of the polymer bubble was accomplished by opening the air valve. The action of the nip rolls not only provided the axial tension but also formed an airtight seal so that a constant pressure could be maintained in the bubble. The nip rolls speed and the amount of air inside the bubble were then simultaneously adjusted to achieve the desired BUR and TUR values. Cooling of the bubble was done by using a single-lip air ring that was located just above the die, directing air at room temperature on the surface of the bubble. The FLH was set up at the desired value by adjusting the cooling air flow rate. However, it was observed that the FLH was also influenced by other variables such as polymer mass flow rate, melt temperature, BUR, and TUR.

The densities of the molten polymers were measured using an Instron capillary rheometer, replacing the capillary by a plug. The description of the method can be found elsewhere (Terry and Yang, 1964). The polymer mass flow rate (w) was measured by weighing the amount of extrudate collected in a known interval of time. This information together with the density of the molten polymer at the die exit (ρ) and the nip rolls speed (V) were used to calculate the TUR as

$$\mathrm{TUR} = \frac{V}{w}\rho\left[\frac{\pi}{4}(D_o^2 - D_i^2)\right], \tag{1}$$

where D_o and D_i are the outer and inner die diameters, respectively.

A video-camera system was used to record the bubble shape and oscillations. The recorded tapes were analyzed by an image analyzer to obtain the bubble diameter and degree of helical instability using the distances of the bubble edges from a reference line at a height well above FLH over a period of time (Figure 2). The concept of the diameter range (D_r), first introduced by Sweeney et al. (1992), was used as a criterion for degree of the helical instability. The average diameter (D_{mean}) and degree of the helical instability (DHI) are then obtained from the following equations:

$$D_{\mathrm{mean}} = P_{l,\mathrm{mean}} - P_{r,\mathrm{mean}} \tag{2}$$

$$D_{\mathrm{max}} = P_{l,\mathrm{max}} - P_{r,\mathrm{min}} \tag{3}$$

$$D_{\mathrm{min}} = P_{l,\mathrm{min}} - P_{r,\mathrm{max}} \tag{4}$$

$$D_r = D_{\mathrm{max}} - D_{\mathrm{min}} \tag{5}$$

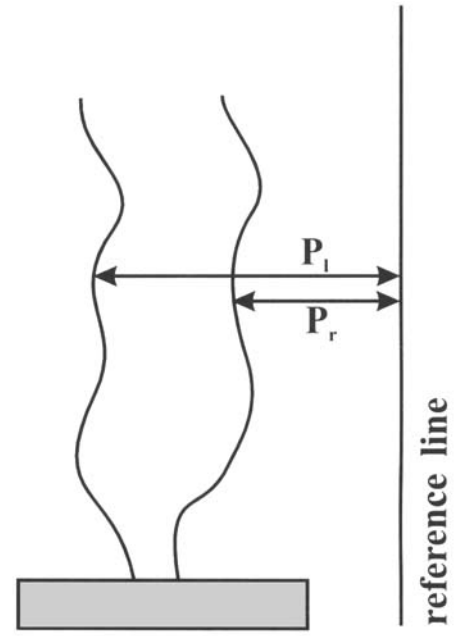

Figure 2. Bubble instability measurement.

$$\mathrm{DHI} = \frac{D_r}{D_{\mathrm{mean}}} \times 100, \tag{6}$$

where P_r is the position of the right bubble edge and P_l the position of the left bubble edge. The maximum, minimum, and mean values were obtained from five measurements. We define a bubble as stable if the degree of instability is less than 20%, partially helical if the degree of instability is between 20 and 40%, and helically unstable if the degree of instability is greater than 40%.

Film blowing experiments were carried out at three different values of TUR and at different FLHs. Due to the limited width of the nip rolls, the maximum attainable BUR with our apparatus was about 5. Operating conditions used for all experiments were within the range of typical industrial conditions.

Results

Rheological measurements

The complex viscosities, η^*, for the four polymers used in this study at their extrusion temperature are shown in Figure 3. At low frequencies, the HDPE has the highest viscosity and the PP, the lowest. The HDPE is more shear-thinning and does not depict a plateau in the low-frequency region. At high angular frequencies, the viscosities of the HDPE and the LLDPE are about the same and those of the LDPE and the PP are identical. The zero shear viscosities (η_0) were determined using the Carreau–Yasuda model (Bird et al., 1987) and are reported in Table 1.

Figure 4 reports data of the storage modulus, G', for the four melts. The most elastic melt is the HDPE, followed by the LLDPE, the LDPE, and the PP. Using these linear viscoelastic data, we can obtain a characteristic relaxation time for the melts, defined by

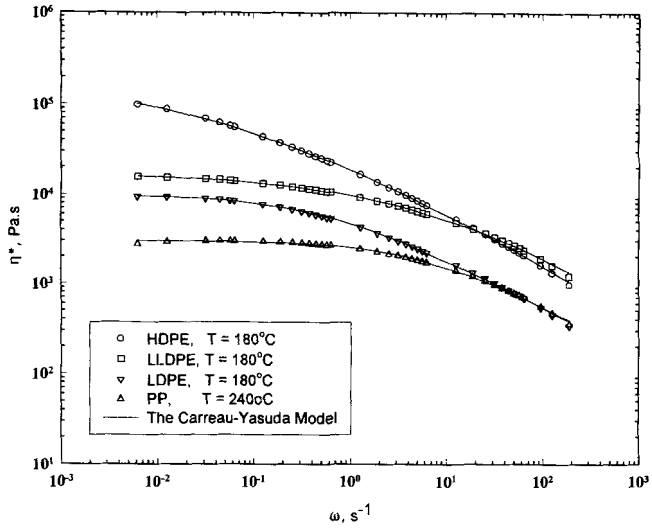

Figure 3. Complex viscosity data vs. frequency for HDPE, LDPE, and LLDPE at 180°C and for PP at 240 °C.

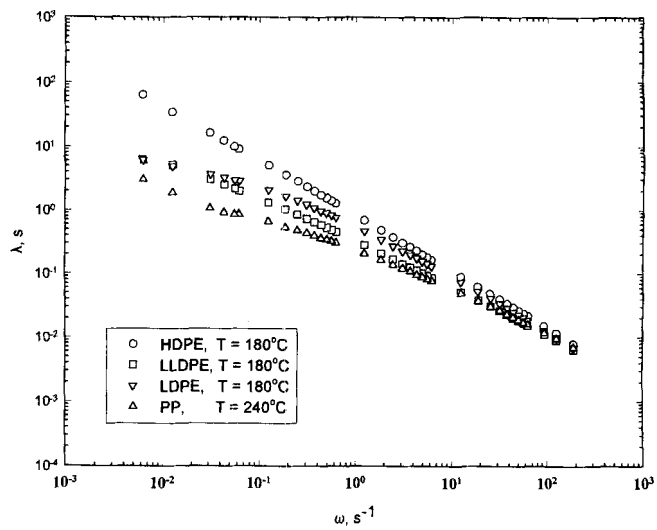

Figure 5. Relaxation time vs. frequency for HDPE, LDPE, and LLDPE at 180°C and for PP at 240°C.

$$\lambda(\omega) = \frac{G'}{G''\omega} \tag{7}$$

and the values are reported in Figure 5. The behavior described by the relaxation times of these melts is quite similar to that of the complex viscosities. At high frequencies the relaxation times of the melts are close to each other. However, at low frequencies, they are quite different; the HDPE has the highest relaxation time and the PP, the lowest, in the same order as the viscosity and the storage modulus. Note that no plateau for λ is observed, indicating that the terminal zone for these polymers has not been reached at the lowest frequencies.

Bubble instabilities

In general, three forms of instabilities and combinations were observed as follows: (1) axisymmetric periodic variations of the bubble diameter; (2) helical motions of the bubble; and (3) variations in the position of the solidification line. We call the first two forms of these instabilities, after previous authors (Han and Park, 1975; Han and Shetty, 1977; Kanai and White, 1984; Minoshima and White, 1986; White and Yamane, 1987), bubble instability and helical instability, respectively. However, we label the third form FLH instability. The term "metastable state" used by the previous authors for such time-dependent oscillations in FLH is misleading.

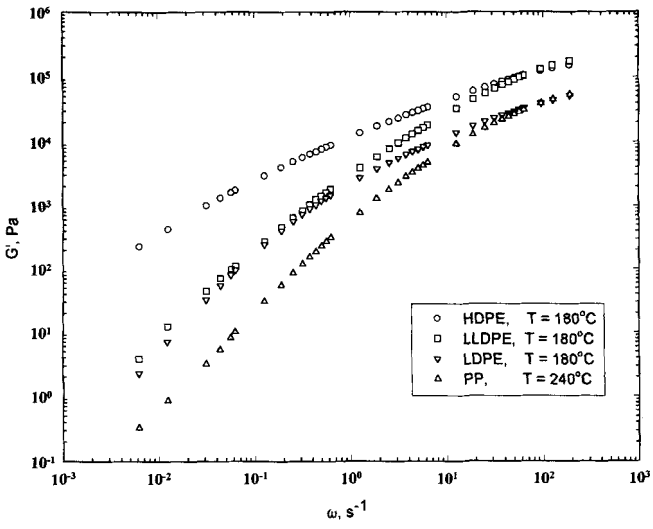

Figure 4. Storage modulus data vs. frequency for HDPE, LDPE, and LLDPE at 180°C and for PP at 240°C.

When the bubble was inflated by a small amount of air, that is, at low BUR, a bubble instability, shown in Figure 6, was observed. This did not appear with the LDPE and the HDPE. No distinct FLH was recognizable and the pressure inside the bubble also fluctuated. The magnitude of the diameter fluctuations increased with time and eventually led to bubble breakage, as already reported by Minoshima and White (1986).

A FLH instability was observed for different operating conditions with the HDPE, the LLDPE, and the PP. Due to the FLH fluctuations, the pressure inside the bubble oscillated and the bubble diameter changed slightly following the FLH fluctuations. For similar conditions Minoshima and

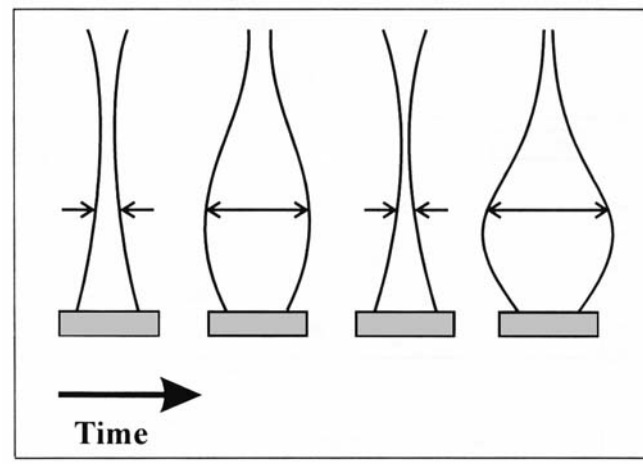

Figure 6. Bubble instability at low BUR.

White (1986) observed fluctuations in the bubble tension, and Fleissner (1988) noticed significant thickness variations in the film.

Helical instability taking place at high BUR was observed with all the polymers. A helical motion develops between the die and the nip rolls, as schematically depicted in Figure 2. In this case, the pressure inside the bubble remained nearly constant.

FLH instability usually grew with time and then combined with helical instability and eventually caused the collapse of the bubble. Helical motion of the bubble normally developed when the FLH moved from the upper limit to the lower one. In the case of the LLDPE and the PP, other types of instabilities were observed leading to the collapse of the bubble.

Stability behavior of LDPE

The instability of the LDPE, the HDPE, the LLDPE, and the PP resins is discussed with the help of diagrams of the FLH vs. the BUR for the three TUR values studied. Figure 7 shows the bubble stability behavior for the LDPE. The symbols on the graphs represent operating conditions. In all cases the bubble is very stable (○ symbols), except for a small unstable space (▽ symbols) at low TUR and intermediate BUR when the FLH is between 150 and 250 mm. As the TUR increases, this unstable space, corresponding to partially helical instability, disappears. Furthermore, at high BUR there is a minimum FLH for which the bubble is stable. Below this FLH value, the bubble is very close to the air ring, and helical motions develop for all of the polymers investigated. In the case of the LDPE this minimum FLH is about 150 mm at a BUR value of 4. This instability may probably be caused by high axial force exerted on the bubble due to the cooling air flow at low FLH. Campbell et al. (1992) have measured the axial force acting on a nondeformable model bubble. They have shown that the air jet produces a significant axial force on the model bubble when the forming region is very close to the air ring.

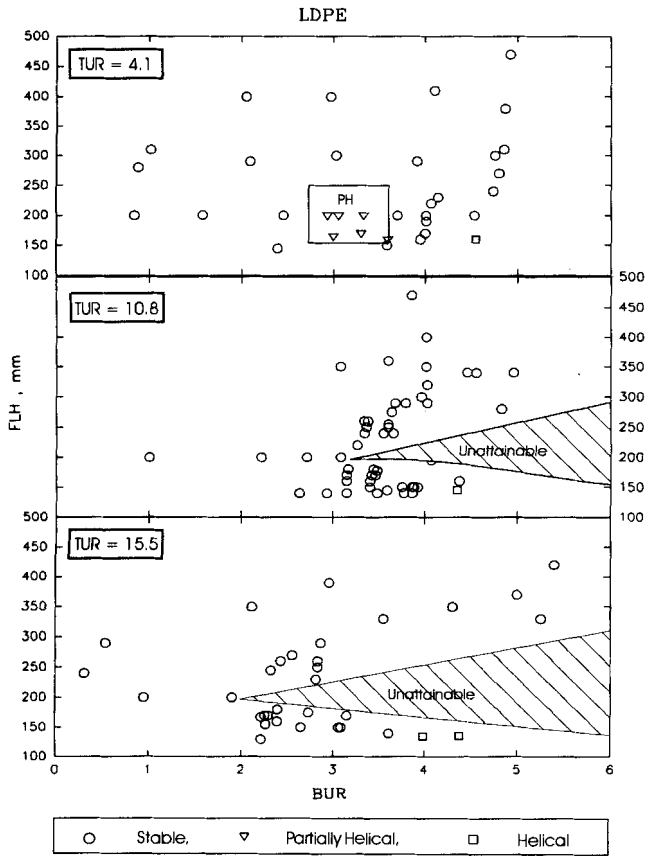

Figure 7. Bubble stability behavior of LDPE.

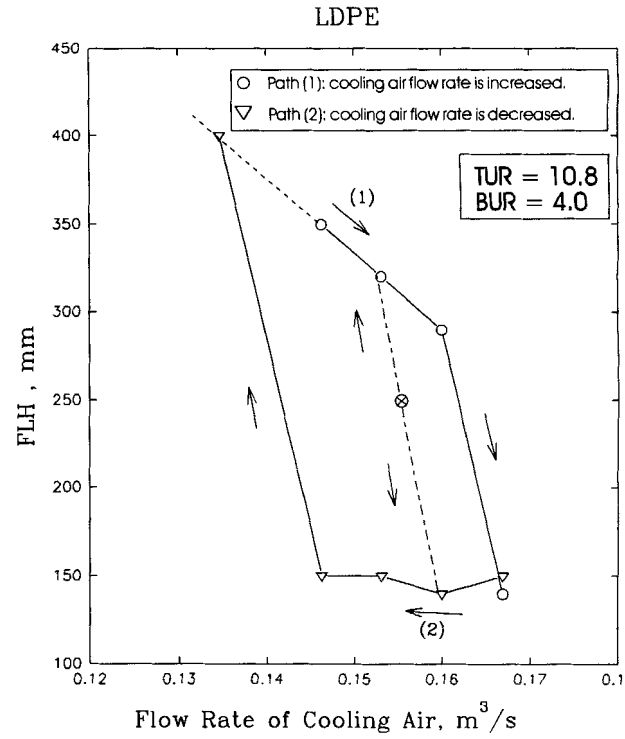

Figure 8. Multiple and nonexisting solutions in film blowing of LDPE at TUR = 10.8 and BUR = 4.0.

As the TUR value increased to 10.8 and 15.5, an interesting phenomenon was observed. As seen in Figure 7 we could not obtain operating points in the hatched area. This area starts from a BUR value of about 3 for TUR = 10.8 and from a BUR value of about 2 for TUR = 15.5 and gets wider when increasing TUR and/or BUR values. In this region, it was observed that a very small change in the cooling air flow rate drastically changed the FLH value and no stable bubble with a FLH value of 200 mm could be obtained. Figure 8 shows the diagram of the FLH vs. the cooling air flow rate at a BUR value of 4.0 and a TUR value of 10.8. The amount of air inside was adjusted to maintain a constant BUR. From this figure we see that as the cooling air flow rate is increased, the FLH value decreases along the upper line (path 1) until a flow rate of 0.160 m^3/s is reached corresponding to a FLH value of 290 mm. At that point any small increase in the air flow rate will bring down the FLH to 150 mm. Then, as the cooling air flow rate is decreased, the FLH slowly increases along the lower line (path 2) until a flow rate of 0.146 m^3/s is reached. Any slight decrease of the air flow rate below this point will bring back the FLH to 400 mm.

Any point inside the space bound by these two steady-state limits represents unstable FLH. For example, if one tries to operate at a FLH equal to 250 mm, as shown in Figure 8 by the cross point, the conditions will be unstable: a pulse in-

crease in the cooling suddenly makes the FLH fall until the FLH reaches 150 mm; on the other hand, a sudden pulse decrease in cooling makes the FLH increase until it reaches 330 mm. These unstable operating points should not be confused with unstable bubble shapes as mentioned before. Furthermore, it is obvious from the figure that at the same cooling air flow rate, two possible FLH values exist. In other words, there are multiple stable steady-state conditions at which film blowing may operate. Multiple steady-state solutions were predicted by Pearson and Petrie (1970) for the isothermal film blowing of a Newtonian liquid. They obtained two BUR values for specified bubble pressure, FLH and TUR. Cain and Denn (1988) have more recently predicted multiple and nonexisting solutions for the film blowing of Newtonian, upper convected Maxwell as well as Marrucci fluids. However, their results were obtained assuming specified values for the take-up force and the bubble pressure (the take-up velocity and the amount of inflating air were controlled in our experiments as in industrial film blowing conditions). The results of Cain and Denn (1988) could not show multiple and nonexisting solutions for these conditions.

Similar results are reported in Figures 9 and 10 for different TUR and BUR values. In these cases the amount of air inside the bubble was kept unchanged, and consequently there were slight BUR changes corresponding to changes in the FLH. At high BUR, the bubble was unstable for very low FLH values, and a stable bubble was obtained at high FLH (see Figure 10).

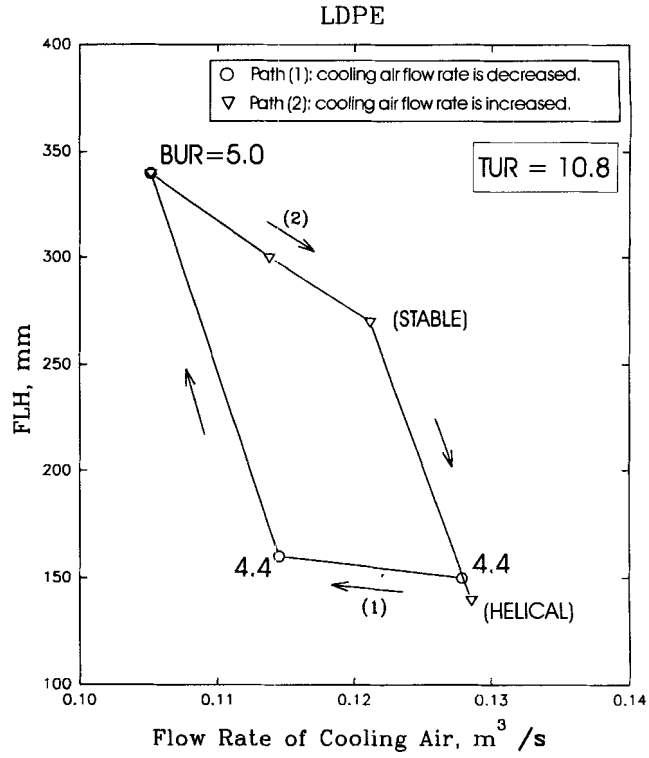

Figure 10. Multiple and nonexisting solutions in film blowing of LDPE at TUR = 10.8.

Numbers represent BUR values.

Stability behavior of HDPE

The stability behavior of the HDPE is shown in Figure 11. One can see that at low TUR value of 4.5 interrelation between BUR and FLH plays an important role in the stability of the bubble. At FLH = 200 mm, the bubble is stable up to a BUR value of about 2.5 (O symbols); afterward, helical instability (▽ and □ symbols) appears. The behavior at FLH = 290 mm is quite different; up to a BUR value of about 2.7 the bubble is stable, then it becomes unstable up to a BUR value of about 3.5, and after that, it becomes stable again. At a FLH value of about 350 mm the bubble is stable up to a BUR value of about 4.2 and then helical instability appears. For FLHs greater than 400 mm, the bubble is stable for the entire experimental range of the BUR.

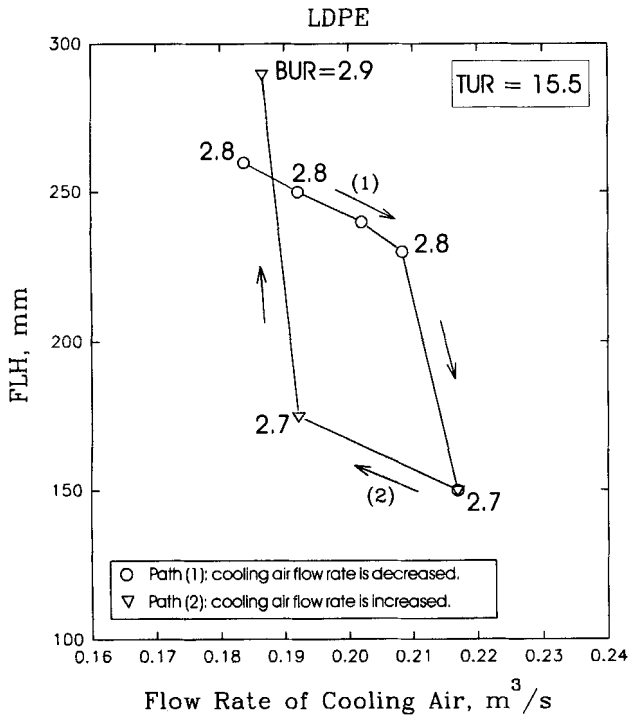

Figure 9. Multiple and nonexisting solutions in film blowing of LDPE at TUR = 15.5.

Numbers represent BUR values.

Figure 11 also shows a region of FLH and helical instabilities at low FLH. As the BUR increases, this unstable space extends to the higher frost line heights. The two regions of helical instability, observed at TUR = 4.5, are totally suppressed at TUR = 16.7. However, the region of FLH and helical instabilities extends to the higher FLHs. No distinct and precise boundaries between stable and unstable regions can be drawn for the TUR values of 11.7 and 16.7. Some stable points overlap or even coincide with unstable points, that is, a switch from stable to unstable state with a slight change in the process conditions. In fact, this phenomenon was observed in a few cases in our experiments; a stable bubble became unstable by disturbing the cooling conditions or the amount of air inside the bubble and the FLH fluctuations increased. However, after a while the fluctuations started to decay and finally a stable bubble was restored. This uncertainty in the boundary between the stable and unstable states was more pronounced at TUR = 16.7, so that a transient space between the two regions is assumed.

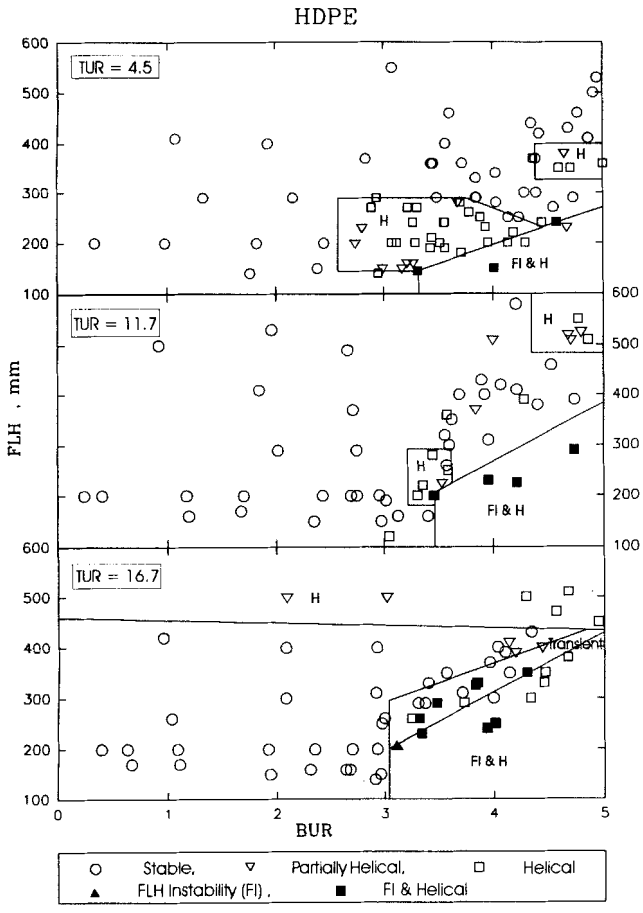

Figure 11. Bubble stability behavior of HDPE.

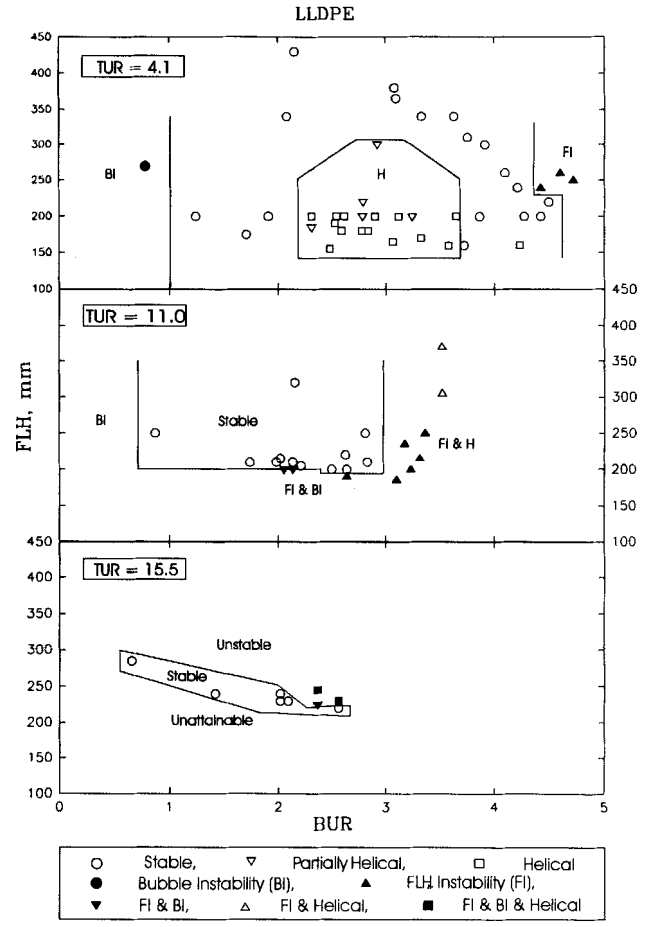

Figure 12. Bubble stability behavior of LLDPE.

Stability behavior of LLDPE

As illustrated in Figure 12, all three forms of instability were observed in film blowing of the LLDPE resin. This polymer is relatively stable at low TUR for a BUR value varying from 1 to 4.5. Below BUR of unity, bubble instability (BI) is observed, as described in Figure 6, and above a BUR value of 4.5, the FLH instability appears. Furthermore, a helically unstable space similar to that observed for the LDPE, but larger, is observed at intermediate BUR values.

The stable operating space is strongly deteriorated by increasing TUR. Although the helically unstable space disappears, the bubble stability is extremely limited by the other types of instabilities. These instabilities are extended to lower BUR so that the maximum attainable BUR at TUR = 15.5 is about 2.6. It also appears that the range of the FLH for which the bubble is stable decreases with increasing TUR. As shown in Figure 12, a region of bubble and FLH instabilities was detected at low FLH. Only a few stable data points were obtained at TUR = 15.5, because of the highly unstable behavior of the LLDPE and the limitations of the cooling system. Decreasing the air flow rate from the maximum amount normally led to highly unstable bubbles. However, bubble instability at low BUR is suppressed with increasing TUR, and the minimum attainable BUR of about 1 at TUR = 4.1 is lowered to about 0.6 at TUR = 15.5. To illustrate the stabilizing effect of increasing TUR, the fluctuations of the pressure inside the bubble were measured and reported in Figure 13. The TUR value was first set at 4.5 (point A) and the bubble was inflated by a small amount of air. Bubble instability was then observed, with periodic fluctuations in the inflation pressure. In a second step at time B, the TUR value was increased to 12.0 and the fluctuations in the pressure reading disappeared, leading to a stable bubble with BUR = 1.1 and FLH = 300 mm. Lastly at time C, the TUR value was changed back to 4.5, causing fluctuations in the inflation pressure to appear slowly and then degenerating in an highly unstable bubble.

Stability behavior of PP

Figure 14 shows that PP has the smallest film blowing operating space among the four polymers investigated. At low values, a stable operating condition requires very low FLH. The bubble is stable in this space for $3.3 \leq \text{BUR} \leq 4.2$. It is expected that the bubble be stable down to a BUR value of 2 even though no data were obtained below BUR = 3.4 because of the limitations of the cooling system. It appears that below a BUR value of 2 the bubble is unstable for the entire range of the FLH. Increasing FLH will eventually cause the bubble to become helically unstable and then show bubble instability.

As TUR is increased, the stable operating space at low FLH disappears. Instead, bubble and FLH instabilities appear as in the case of the LLDPE and the HDPE. This unsta-

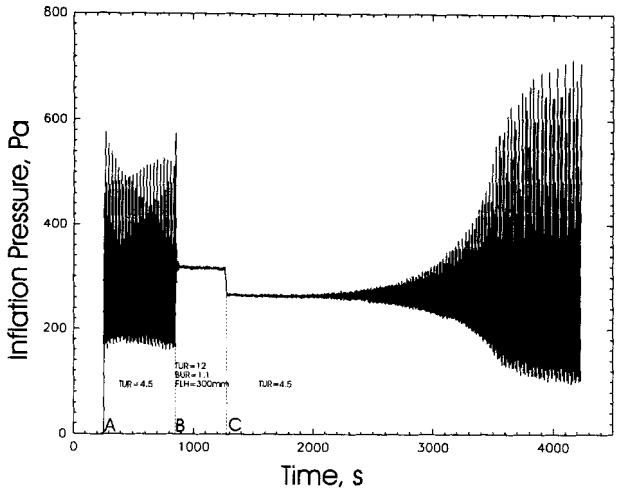

Figure 13. Bubble instability of LLDPE at low BUR.

ble space extends to the higher FLH values as the TUR value is increased. However, a small stable operating space appears at higher FLH. This space is restricted to BUR between 1 and 2 at TUR = 10.8 and a maximum BUR value of 2.2 is obtained for TUR = 15.5. Increasing TUR has a stabilizing effect on the bubble at low BUR, as observed for the LLDPE.

Discussions and Conclusions

The LDPE was found to have the largest operating space, whereas the PP had the smallest one. For the four polymers studied the relative order of stability is as follows:

$$\text{LDPE} > \text{HDPE} > \text{LLDPE} > \text{PP}.$$

As far as we are aware there are no data in the literature on the stability of PP. For the other polymers, our results are in agreement with the previously published results. Comparing the stability order of these four polymers, it is obvious that bubble instabilities cannot be correlated with the simple shear rheological data. We recall that it is virtually impossible to predict the extensional properties from simple shear data: film blowing is controlled by the extensional flow behavior of the material and coupling effects between heat transfer and rheological properties. The more stable behavior of the LDPE is attributed to its strain hardening in elongational flow, as reported by Kanai and White (1984) and White and Yamane (1987). The elongational behavior of the other three polymers is not as well characterized, although there are indications that linear PEs and PP are strain thinning (Kanai and White, 1984; Hingmann and Marczinke, 1994). Strain-hardening elongational properties have been reported for a HDPE at low elongational rates (Fleissner, 1988). The HDPE used in our experiments is the second most stable polymer. Obviously, more work is needed to clarify the importance of strain hardening coupled with cooling effects on film blowing stability.

Regions of multiple and nonexisting solutions were detected for the LDPE as the TUR value was increased; a slight change in the cooling air-flow rate would increase the FLH value to a upper steady state or decrease it to a lower steady state. Bubble instability at low BUR was observed with the

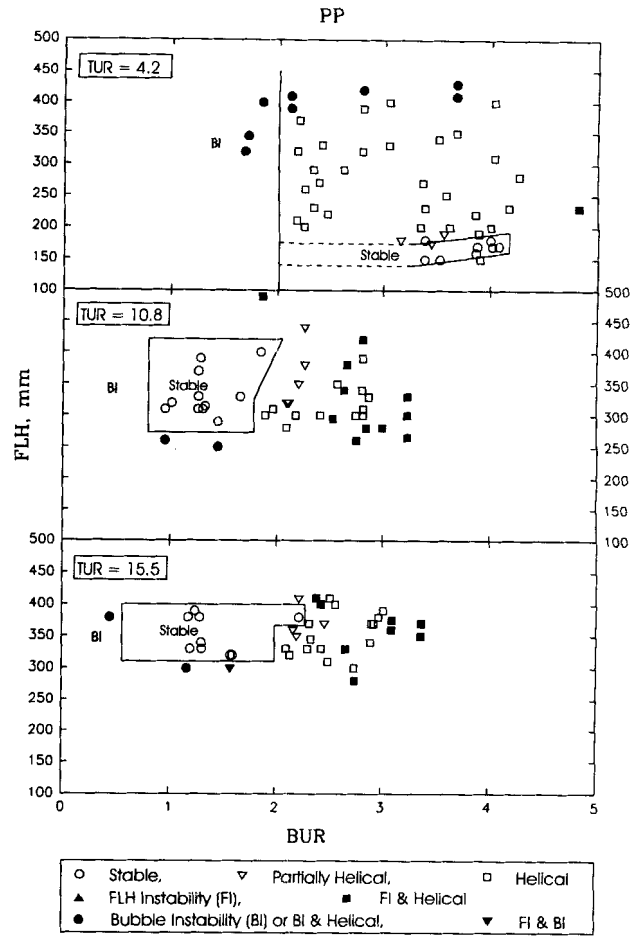

Figure 14. Bubble stability behavior of PP.

LLDPE and the PP. The mechanism of bubble instability is believed to be primarily rheological in character. Die exit effects such as extrudate swelling as well as extensional properties may play an important role on bubble stability. As the BUR and/or the TUR are increased exit effects become negligible and bubble stability is increased.

For the HDPE, the LLDPE, and the PP the instability of the film blowing process (mainly FLH instability) increased with increasing BUR and TUR values and decreasing FLH values (the effect of decreasing FLH on the stability was clearly seen at high TUR). Interestingly, the stretch rate in this process, as measured by Kanai and White (1984) for HDPE, LDPE, and LLDPE, follows the same pattern, that is, it increases with increasing BUR, decreasing FLH, and increasing TUR. Therefore, we speculate that as the stretch rate (and, hence, the tensile stress) increases and reaches a critical value, instability phenomena in film blowing start to appear. We assume that at critical stretch rate a local thinning of the tubular film may happen, resulting in the instability of the bubble by a mechanism proposed by Fleissner (1988). This is briefly described below.

Because of the local thinning of the tubular film in the neck zone, which is accompanied by a decrease in the drawing force, melt is accumulated in the die region since both mass-flow rate of polymer and take-up velocity are constant. However, the thinner section can be drawn more easily and

cooling is more rapid, resulting in a shift down of the FLH. Meanwhile, mass accumulation inevitably causes a thickening of the film. The thicker film can no longer be easily stretched, resulting in a higher tensile force and therefore the upward shift of the FLH. The higher tensile stress will again thin down the tubular film in the die region and this cycle will continue and finally cause the collapse of the bubble.

Helical instability was observed with all the polymers. It usually decreased with increasing TUR and it also depended on the bubble shape. Air drag forces play an important role on the helical instability. At low TUR values, the air drag forces are comparable to the viscous forces. As a consequence, the bubble is sensitive to the drag forces. As the drag forces can vary substantially along the length of the bubble, this may cause the forces acting on the bubble to be unbalanced, and consequently lead to helical instability. With increasing TUR, the viscous forces become predominant and this makes the bubble less sensitive to the drag forces.

Finally, the bubble was found to be more stable as the FLH value was increased, except in the case of the PP at low TUR. This is clearly in disagreement with the suggestion of Minoshima and White (1986), who have stated that increasing the FLH value decreases the range of stable conditions. However, our results show an upper limit above which the bubble becomes unstable.

We finally discuss our observations on bubble stability in light of the theoretical predictions of Cain and Denn (1988), who analyzed bubble stability to infinitesimal disturbances. As mentioned in a previous paragraph, helical instability can be attributed to the existence of air drag forces. However, this term was neglected in the analysis of Cain and Denn. Therefore, it should not be expected that helical instability be predicted by their analysis. For the conditions of fixed take-up velocity and constant amount of inflating air, they predicted the occurrence of instability at very low BUR (about 0.3) for a Newtonian fluid. Instability in film blowing of a Maxwell fluid was also predicted at very low BUR, but confined to a smaller region. This instability was totally suppressed with increasing relaxation time. This is qualitatively in agreement with our experimental observations. The HDPE has a higher relaxation time than the LLDPE, as shown in Figure 5. Figure 11 shows that bubble instability at low BUR was not observed with the HDPE, in agreement with the model predictions. On the other hand, Cain and Denn's analysis does not provide any realistic information on the occurrence of instability at high BUR. They have predicted instability at high BUR to occur at a thickness reduction in excess of 700 for a Newtonian fluid and at a thickness reduction of about 230 for a Maxwell fluid. Increasing relaxation time did not change significantly the thickness reduction at which instability at high BUR appeared. In contrast, we observed instabilities at high BUR (FLH instability) for the HDPE and the LLDPE at thickness reductions as low as about 20. In summary, our experimental results are in little agreement with the predictions of Cain and Denn (1988). This stresses the need for a more realistic rheological constitutive equation, with parameters characterizing the elongational properties of polymers, and the necessity of heat-transfer considerations in the analysis of the film blowing process. In a forthcoming article we will examine the elongational flow behavior of molten polymers in film blowing using a birefringence method.

Acknowledgment

The authors wish to acknowledge financial support received from the FCAR programme of the Province of Quebec and from NSERC. We are also thankful to the anonymous reviewers for their helpful comments.

Notation

M_n = number-average molecular weight, kg/kmol
M_w = weight-average molecular weight, kg/kmol
M_z = z-average molecular weight, kg/kmol
ω = frequency, rad/s

Literature Cited

Ast, W., "Air Cooling on Blown Film Lines," *Kunstoffe*, **64**, 146 (1974).
Bird, R. B., R. C. Armstrong, and O. Hassager, *Dynamics of Polymeric Liquids*, 2nd ed., Wiley-Interscience, New York (1987).
Cain, J. J., and M. M. Denn, "Multiplicities and Instabilities in Film Blowing," *Poly. Eng. Sci.*, **28**, 1527 (1988).
Campbell, G. A., N. T. Obot, and B. Cao, "Aerodynamics in the Blown Film Process," *Poly. Eng. Sci.*, **32**, 751 (1992).
Fleissner, M., "Elongational Flow of HDPE Samples and Bubble Instability in Film Blowing," *Int. Poly. Process.*, **2**, 229 (1988).
Ghijsels, A., J. J. S. M. Ente, and J. Raadsen, "Melt Strength Behavior of PE and its Relation to Bubble Stability in Film Blowing," *Int. Poly. Process.*, **5**, 284 (1990).
Han, C. D., and J. Y. Park, "Studies on Blown Film Extrusion: III. Bubble Instability," *J. Appl. Poly. Sci.*, **19**, 3291 (1975).
Han, C. D. and R. Shetty, "Flow Instability in Tubular Film Blowing: 1. Experimental Study," *IEC Fundam.*, **16**, 49 (1977).
Hingmann, R., and B. L. Marczinke, "Shear and Elongational Flow Properties of Polypropylene Melts," *J. Rheol.*, **38**, 573 (1994).
Kanai, T., and J. L. White, "Kinematics, Dynamics and Stability of the Tubular Film Extrusion of Various Polyethylenes," *Poly. Eng. Sci.*, **24**, 1185 (1984).
Minoshima, W., and J. L. White, "Instability Phenomena in Tubular Film, and Melt Spinning of Rheologically Characterized High Density, Low Density and Linear Low Density Polyethylenes," *J. Non-Newt. Fluid Mech.*, **19**, 275 (1986).
Obijeski, T. J., and K. R. Pruitt, "Improving the Output and Bubble Stability of Thick Gauge Blown Film," *ANTEC 92*, p. 150 (1992).
Pearson, J. R. A., and C. J. S. Petrie, "A Fluid-Mechanical Analysis of the Film-Blowing Process," *Plastics Poly.*, **38**, 85 (1970).
Sweeney, P. A., G. A. Campbell, and F. A. Feeney, "Real Time Video Techniques in the Analysis of Blown Film Instability," *Int. Poly. Process*, **7**, 229 (1992).
Sweeney, P. A., and G. A. Campbell, "Blown Film Stability," *ANTEC 93*, p. 461 (1993).
Terry, B. W., and K. Yang, "A New Method for Determining Melt Density as a Function of Pressure and Temperature," *SPE J.*, **20**, 540 (1964).
White, J. L., and H. Yamane, "A Collaborative Study of the Stability of Extrusion, Melt Spinning and Tubular Film Extrusion of Some High-, Low- and Linear-Low Density Polyethylene Samples," *Pure Appl. Chem.*, **59**, 193 (1987).
Yeow, Y. L., "Stability of Tubular Film Flow: A Model of the Film-Blowing Process," *J. Fluid Mech.*, **75**, 577 (1976).

Manuscript received May 11, 1995, and revision received Aug. 16, 1995.