# Fundamental Foaming Mechanisms Governing the Volume Expansion of Extruded Polypropylene Foams

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**ABSTRACT:** This article describes the fundamental foaming mechanisms that governed the volume expansion behavior of extruded polypropylene (PP) foams. A careful analysis of extended experimental results indicated that the final volume expansion ratio of the extruded PP foams blown with butane was governed by either the loss of the blowing agent or the crystallization of the polymer matrix. A charge coupling device (CCD) camera was installed at the die exit to carefully monitor the shape of the extruded PP foams. The CCD images were analyzed to illustrate both mechanisms, gas loss and crystallization, during foaming at various temperatures, and the maximum expansion ratio was achieved when the governing mechanism was changed from one to the other. In general, the gas loss mode was dominant at high temperatures and the crystallization mode was dominant at low temperatures. When the gas loss mode was dominant, the volume expansion ratio increased with decreasing temperature because of the reduced amount of gas lost. By contrast, when the crystallization mode was dominant, the expansion ratio increased with increasing temperature because of the delayed solidification of the polymer. The processing window variation with the butane concentration, the change in the temperature ranges for the two governing modes, and the sensitivity of melt temperature variations to the volume expansion ratio are discussed in detail on the basis of the obtained experimental results for both branched and linear PP materials. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 91: 2661–2668, 2004

**Key words:** foam extrusion; poly(propylene) (PP); blowing agents; diffusion; crystallization

## INTRODUCTION

Thermoplastic foams possess a cellular structure created by the expansion of a blowing agent. This internal structure provides unique properties that enable the foamed plastics to be used effectively in various industrial applications.[1] Because of its outstanding functional characteristics and low material cost, polypropylene (PP) foams have been considered as a substitute for other thermoplastic foams in industrial applications. They are a member of the semicrystalline polyolefin family, which is resistant to chemicals and abrasion. They have a number of advantages over polystyrene and polyethylene.[2] They have a higher rigidity compared to other polyolefins, they offer a higher strength than polyethylene and a better impact strength than polystyrene, and they provide a higher service temperature range and a good temperature stability. However, only limited research has been conducted on the production of PP foams because of their weak melt strength and melt elasticity, which make comparison of the foam to that of other plastics difficult.[3]

Most polyolefin foams are produced by the expansion process, which is based on the expansion of a gaseous phase dispersed throughout the polymer melt. The gaseous phase may be generated by the separation of a dissolved gas, vaporization of a volatile liquid, or release of gas from a chemical reaction. Regardless of the type of blowing agent, the expansion process comprises three major steps: nucleation, bubble growth, and stabilization. Nucleation, or the formation of expandable bubbles, can begin within a polymer melt that has been supersaturated with the blowing agent. Once nucleated, a bubble continues to grow as the blowing agent diffuses into it. This growth will continue until the bubble stabilizes or ruptures.[4]

When cells are nucleated, the foam density decreases as the available blowing agent molecules diffuse into the cells. The growth rate of the cells is limited by the diffusion rate and the stiffness of the viscoelastic polymer/gas solution. In general, cell growth is affected primarily by the time allowed for the cells to grow, the temperature of the system, the state of supersaturation, the hydrostatic pressure or stress applied to the polymer matrix, and the viscoelastic properties of the polymer/gas solution.[5,6]

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During the extrusion of PP foams, the blowing agent that promotes foam expansion may escape through the exterior skin of the foam, resulting in the contraction of the foam.[7–10] Because the expansion of foam relies on the presence of the blowing agent within the foam, it is desirable to devise a means for preventing gas loss during foaming to achieve a large volume expansion ratio.

There are few studies that have investigated the foaming behavior of PP. Propylene materials possess a weak melt strength, which increases the difficulties of foaming compared to other plastics. When the melt strength is too weak, the cell walls separating the bubbles may not have enough strength to bear the extensional force and may rupture very easily during foaming. As a result, foamed PP products usually have a high open-cell content and, thus, are unsatisfactory for many applications.

To improve the foamability and thermoformability, material modification or new resin developments have been conducted for PP materials. Earlier efforts have included the crosslinking of PP resins; significantly improved volume expandability, cell uniformity, and thermoformability of the foams were observed.[11–14] Efforts have also been made to promote long-chain branching[15–19] to improve the melt strength of PP material. Much improved foamability and thermoformability were observed from long-chain, branched, high-melt-strength (HMS) PP resins.[3,16,20,21] Various other grades of HMS PP resins[22–24] have also been presented.

The foaming behaviors of various PP resins have been investigated. The cell nucleating behavior and the final cell density of extruded PP foams with various nucleating and blowing agents have been studied extensively for fine-celled PP foam applications.[3,25–30] The volume expansion behavior and the final foam density have also been studied extensively for low-density PP foam applications.[31–36]

This article presents a qualitative modeling for the volume expansion behavior. The fundamental mechanisms governing the volume expansion of PP foams are identified first, and the volume expansion phenomena are described on the basis of our experimental observation. The system setup used for monitoring the expansion phenomena of extruded foam is then described. The procedure for monitoring the expansion mechanisms and the image analysis are elucidated. Consequently, the effects of processing parameters on the volume expansion behavior are depicted on the basis of the extrudate images.

## FUNDAMENTAL MECHANISMS GOVERNING THE VOLUME EXPANSION OF PP FOAMS

A careful analysis of extended results obtained at various processing conditions indicated that the final volume expansion ratio of the extruded PP foams blown with butane was governed either by the loss of blowing agent through the foam skin or by the crystallization of the polymer matrix. In general, on exiting the die, the foaming extrudate exhibited one of the shapes depicted in Figure 1, depending on the temperature of the extrudate. At higher temperatures, the cross-section of the extrudate expanded suddenly; that is, it had a higher initial angle ($\theta$), and this angle decreased as the processing temperature decreased. Below an optimum processing temperature, the foaming of extrudate was inhibited, and $\theta$ was substantially reduced. The mechanisms governing the volume expansion of PP foams are explained in the following sections.

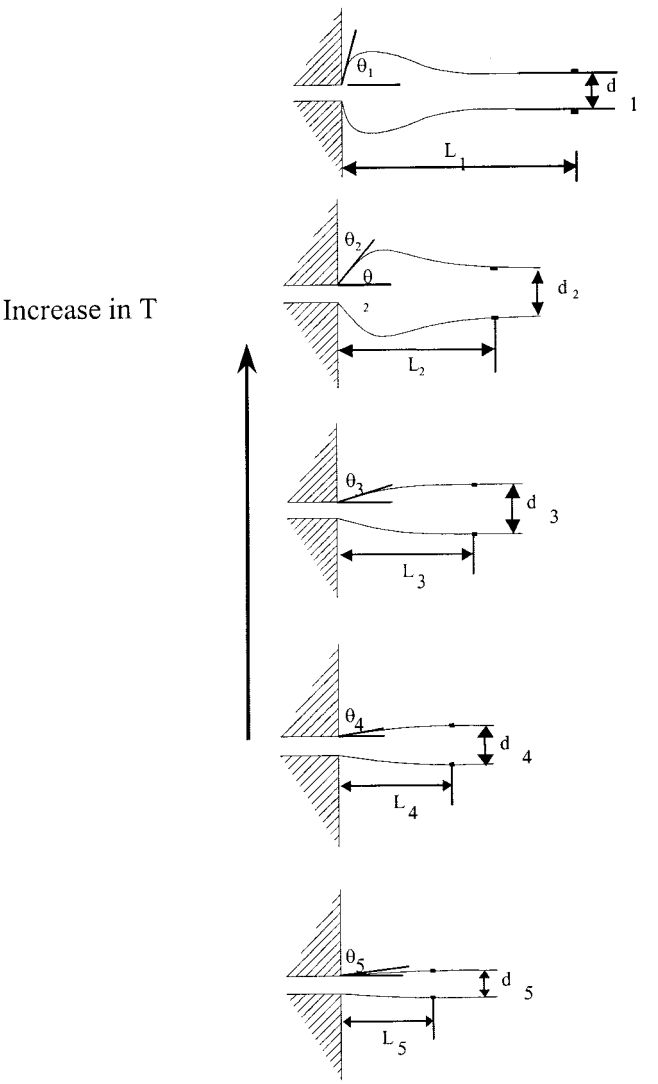


**Figure 1** Effect of processing temperature on the extrudate shape.

### Gas loss

The gas loss phenomena that occurred during foam processing could be correlated with the melt temper-

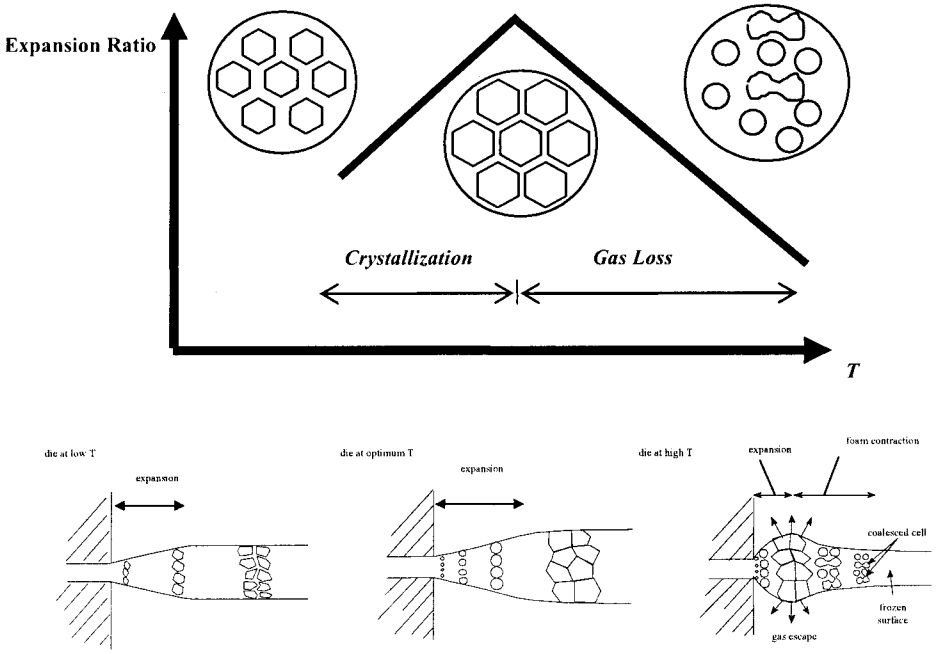


**Figure 2** Effect of gas loss and crystallization on the volume expansion.

ature. The diffusivity of the blowing agents at elevated temperatures was very high; therefore, if the processing temperature was too high, the gas could easily escape from the extruded foam because of its higher diffusivity at elevated temperatures. In addition, as the cell expansion increased, the cell wall thickness decreased, and the resulting rate of gas diffusion between cells increased. Consequently, the rate of gas escape from the foam to the environment increased. Gas escape through the thin cell walls decreased the amount of gas available for the growth of cells, resulting in a lowered expansion. Moreover, if the cells did not freeze quickly enough, they tended to shrink because of loss of gas through foam skin causing overall foam contraction. This mechanism is schematically shown in Figure 2.

This phenomenon of gas escape at high temperatures and the resultant foam contraction during the cell growth stage at elevated temperatures were observed during experimentation. The extruded foam initially expanded as the nucleated cells grew very fast and then eventually contracted. The maximum expansion occurred very close to the die exit, with the diameter of the initially expanded foam being larger than that of the final foam extrudate. This indicated that the volume expansion ratio of the initially expanded state was considerably high, and therefore, the shapes of expanded cells at this state were not spherical but polyhedral with thin cell walls (Fig. 2).

## Crystallization

The crystallization behavior of semicrystalline materials is another critical factor that affects the maximum expansion ratio in plastic foam processing. For semicrystalline polymers, the polymer melt solidifies at the moment of crystallization during cooling. Therefore, in the foam processing of PP, the foam structure froze at the crystallization temperature during the foaming process. If the crystallization occurred in the primitive stage of foaming, that is, before the dissolved blowing agent fully diffused out of the plastic matrix and in the nucleated cells, the foam cannot be fully expanded. Therefore, to achieve the maximum volume expansion ratio from the PP foam, the crystallization (or solidification) should not occur before all the dissolved gas diffuses out into the nucleated cells. On exiting the die, the temperature of the melt decreased because of external cooling outside the die and the cooling effect due to the isentropic expansion of the blowing gases. Thus, the processing temperature at the die determined the time after which the polymer melt solidified. Therefore, to give enough time for the gas to diffuse into the polymer matrix, the processing temperature should be high enough. If the processing temperature was too close to the crystallization temperature, the polymer melt solidified too quickly before the foam was expanded fully, as shown in the initial section of Figure 2.

However, if the temperature was too high, the solidification time was too long, and the gas that had

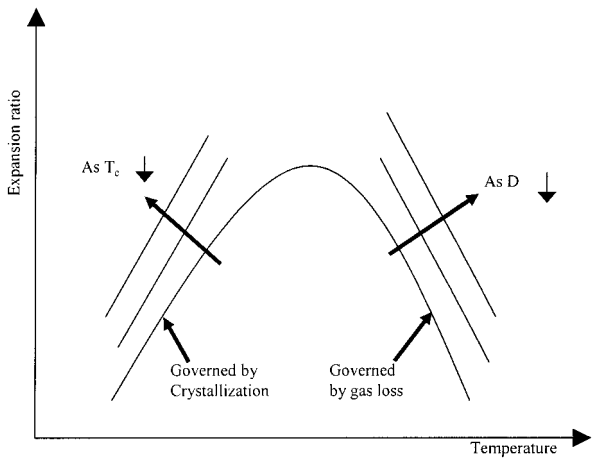


**Figure 3** Fundamental volume expansion mechanism of PP foams.

diffused out of the plastic melt to the nucleated cells escaped out of the foam, as discussed previously. This indicated that there was an optimum processing temperature for the achievement maximum expansion, as shown in the middle section of Figure 2. If the melt temperature (i.e., the processing temperature) was too high, the maximum volume expansion ratio was governed by gas loss, and the volume expansion ratio increased as the processing temperature decreased. If the processing temperature was too low, the volume expansion ratio was governed by the solidification (i.e., the crystallization) of PP, and the volume expansion ratio increased as the temperature increased.

In addition to the effects of the processing parameters on the crystallization, the foaming additives and materials parameters also contributed to changes in crystallization temperatures. The effect of these parameters on the volume expansion ratio is shown in Figure 3.

## EXPERIMENTAL

### Materials

The material used in this study was commercial PP resin supplied by Borealis GmbH (Linz, Austria): Daploy WB130HMS as an HMS branched PP resin. The MFR (ISO 1133, 230°C/2.16 kg) of this material was 2.3 dg/min. Talc A7 with a particle size of less than 7 $\mu$m was used as the nucleating agent (Naintsch, Austria). *N*-Butane was used as a blowing agent (Matheson Gas).

### Setup

Figure 4 shows a schematic of the tandem foam extrusion system used in our study. It consisted of a 5-hp extruder driver with a speed control gearbox (Brabender, Prep Center); a first 0.75-in. extruder (Brabender, 05-25-000) with a mixing screw (Brabender, 05-00-144) with a length/diameter ($L/D$) ratio of 30:1; a second 1.5-in. extruder (Killion, KN-150) with a built-in 15-hp variable speed drive unit with a $L/D$ ratio of 18:1; a positive displacement pump for the injection of the blowing agent to the polymer melt; a static mixer (Omega, FMX-84441-S) as a diffusion-enhancing device; a gear pump (Zenith, PEP-II 1.2 cc/rev), where the volumetric displacement was properly controlled by the motor; a heat exchanger for the cooling of the polymer melt, which contained homogenizing static mixers (Labcore, model H-04669-12);

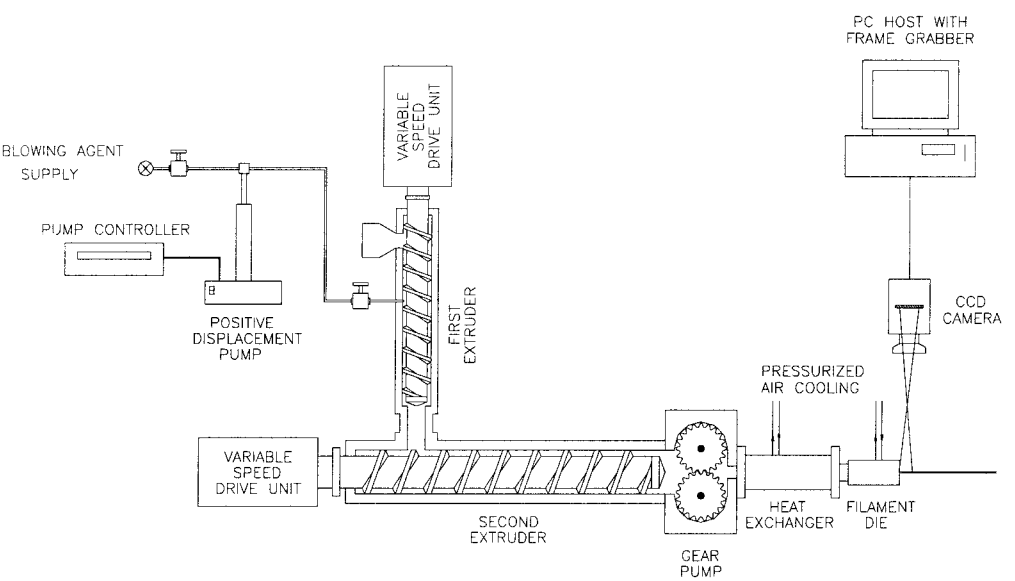


**Figure 4** Experimental setup.

and a cooling sleeve for the precise control of the die temperature. The first extruder was used for the plastication of the polymer resin. The second extruder provided mixing and initial cooling for the polymer melt. The gear pump controlled the polymer melt flow rate, independent of temperature and pressure changes. The heat exchanger provided further cooling and homogenization of melt temperature. A filamentary die was used in this study. An online progressive scan imaging system was mounted at the die exit to capture images of the extrudate coming out of the die, with a frame grabber and image processing software. The progressive scan imaging system consisted of a charge coupling device (CCD) camera (CV M10), which had a very high shutter speed up to 1/80,000 s, with a magnifying lens (Navitar), a frame grabber (PC Vision), and image processing software (Sherlock).

### Procedure

The PP materials pellets, mixed with 0.8 wt % talc, were first fed into the barrel through the hopper and were completely melted by the screw motion. A metered amount of blowing agent was then injected into the extrusion barrel by a positive displacement pump at a given weight percentage and mixed with the polymer melt stream. When the gas was injected into the extrusion barrel, the remaining section of the first screw and the second screw generated a shear field to completely dissolve the gas in the polymer melt via convective diffusion.[37] The single-phase polymer/gas solution went through the gear pump and was fed into the heat exchanger, where it was cooled to a designated temperature. The cooled polymer/gas solution entered the die, and foaming occurred at the die exit. The melt and die temperatures were synchronized for simplicity in this study. With all of the other materials and processing parameters, such as screw speed, gear pump speed, blowing agent content, and barrel temperatures, fixed, the melt and die temperatures were lowered in stages, and samples were randomly collected at each designated temperature only after there was no change observed in the pressure. The foamed extrudate was monitored while the processing temperatures were changed.

The foam samples were characterized with an optical microscope (Wild Heerburgg) and/or a scanning electron microscope (Hitachi 510). The foam samples were dipped in liquid nitrogen and were then fractured to expose their cellular morphology before the characterization of foam structure.

## RESULTS AND DISCUSSION

### Effect of processing conditions on volume expansion and cell density

Figure 5(a) depicts the expansion ratio versus the melt temperature for the branched PP materials. Equivalently, the blowing agent efficiency is plotted against the melt temperature in Figure 5(b). The volume expansion ratio was a sensitive function of the melt temperature. When the melt temperature was as high as 180°C, the achieved volume expansion ratio was only about 2 times. This means that when the melt temperature was too high, most of the gas escaped through the hot skin layer of foam during expansion because of the high diffusivity of the gas.[8] However, when the temperature of the polymer melt was lowered, a high volume expansion ratio was achieved. In other words, the amount of gas lost was reduced because of the lowered diffusivity, and more gas remained in the foam, resulting in a greater volume expansion at the lower melt temperature.

There was an optimum temperature that produced the maximum expansion ratio. When the temperature was higher than this temperature, the volume expansion ratio increased as the temperature decreased. This implies that the volume expansion behavior was limited by the gas loss phenomena in this temperature range. By contrast, when the temperature was below the temperature at which the maximum volume expansion occurred, the volume expansion ratio decreased as the temperature decreased. The early crystallization of the PP materials when the processing temperature was too low must have been the reason for the lower expansion ratio as discussed previously.

Figure 5(c) depicts the cell density versus the melt temperature for the branched PP materials. The figure shows that the cell density did not change when the temperature of the melt was varied. Because the same die was used in all of the experiments, the pressure built by the resistance of the die was varied as a function of the melt temperatures, as shown in Figure 5(d). This implied that the variation of the melt pressure had no major effect on the cell density. It seemed that this was due to the high solubility of butane in the polymer. Although, there is no specific data available in the literature about the solubility of butane in PP at high pressures and high temperatures, the solubility of halogenized hydrocarbon (FC 11) in PP was reported to be as high as 90 wt % at 13.8 MPa and 200°C.[38] We, therefore, expected that the solubility of butane in PP would be also high. The amount of injected butane in the range of 5–20 wt % must have been far below the solubility limit under the processing conditions, and the entire amount of injected butane was believed to dissolve in the polymer melt regardless of the processing pressure. In other words, the thermodynamic instability induced from the dissolved butane and PP matrix was not affected by the processing pressure. As a result, the driving force to nucleate bubbles did not vary as the processing pressure was increased. It appeared that cell density was mainly determined by the talc content[30] and the amount of butane.

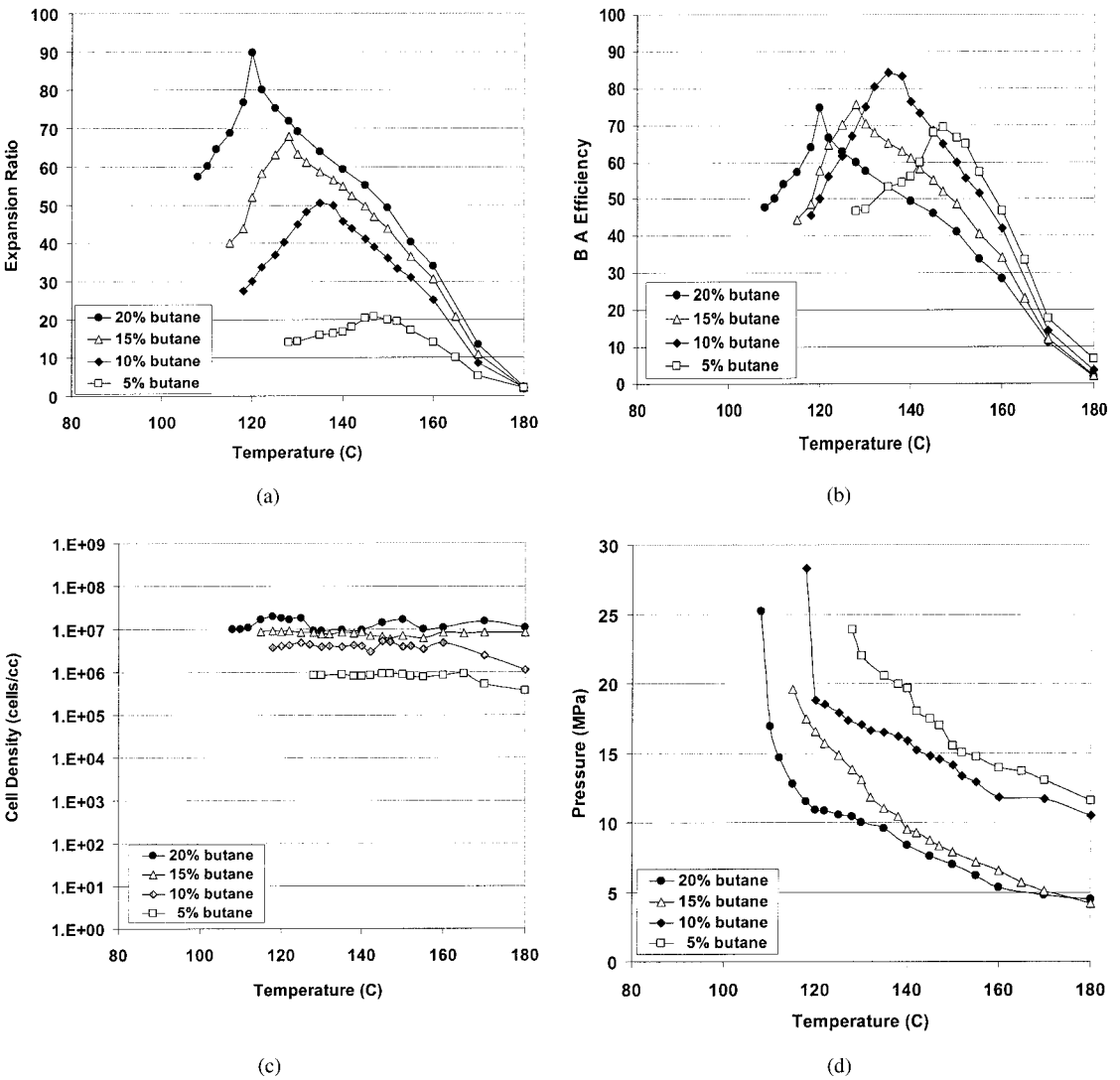


**Figure 5** (a) Expansion ratio of branched PP, (b) blowing agent efficiency, (c) cell densities of branched PP, and (d) die pressure profile.

### Visualization of expansion behavior with a CCD camera

From the captured images, the angle of initial swelling of the foamed polymer and the extrudate diameter as a function of the distance from the die exit were measured. The images of the foamed extrudate were analyzed and the effects of gas loss and crystallization were extracted. Some sample images are shown in Figure 6. The sample results for the branched PP material with a 15% butane concentration at three different processing temperatures are elucidated in the following sections.

Figure 7 shows the effect of processing temperature on the initial volume expansion behavior characterized by $\theta$ of the extrudate. The figure shows that at higher temperatures, the initial expansion rate and, consequently, $\theta$ were quite significant. However, because gas loss was accelerated at this high temperature, the final diameter ($d$) of the extruded foam was small. As the temperature decreased, the initial expansion rate ($\theta$) decreased, and $d$ of the extruded foams increased because of the decreased amount of gas loss. There existed an optimum temperature for the achievement the maximum diameter of the extrudate. When the temperature was further decreased, and because of the early crystallization, the foamed extrudate was frozen before the extrudate was fully expanded. As a consequence, $d$ was small. The results are summarized in Figure 8.

Figure 9 shows the changes occurring in the extrudate diameter, as the distance from the die exit increased. At the optimum die temperature of 130°C, the extrudate diameter reached its maximum value of 5 mm at a distance of 5 mm away from the die exit. At this point the foam extrudate reached the crystallization temperature and retained its shape due to the

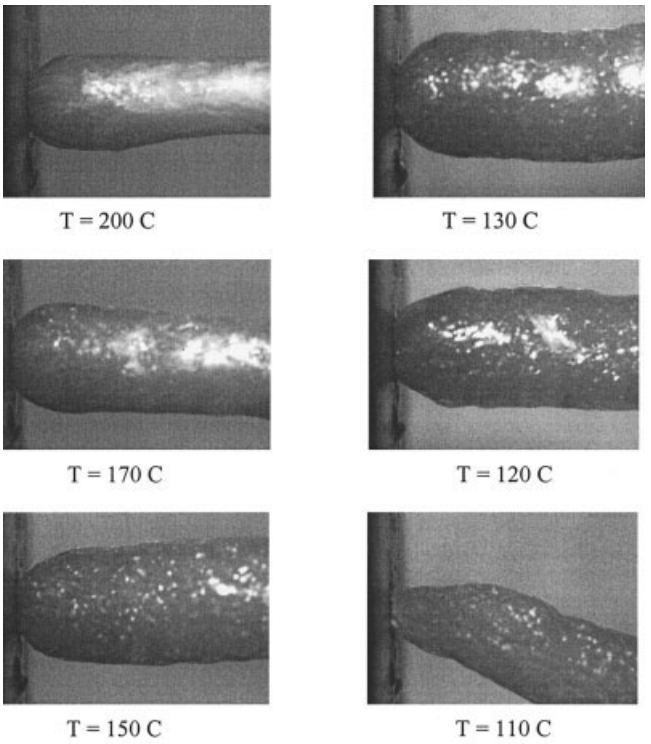


**Figure 6** Images of the foam extrudate coming out of the die.

solidification of the polymer. At the lower temperature of 120°C, the polymer reached the crystallization temperature earlier, about 3 mm away from the die exit, and froze at this point. Thus, at the same gas content of 15%, the maximum diameter achieved was about 4.6 mm, instead of 5 mm, which resulted in the sacrifice of the expansion ratio. In the meanwhile, at a higher temperature of 200°C, the effect of gas loss was more pronounced. First, the gas at a higher temperature caused a sharper initial expansion ratio, and the rate of diameter growth was greater. Simultaneously, the gas loss due to the increased diffusivity of the gas resulted in a lowered maximum diameter in the extrudate, which was about 3.6 mm and was observed about 2.5 mm away from the die exit. As the temperature of the extrudate was still above the crystallization temperature of the polymer, further gas loss occurred, and the diameter continued to decrease beyond the range of the photographic equipment. This decrease in diameter could attributed to either contraction due to cooling or additional gas loss by diffusion through the foam skin.

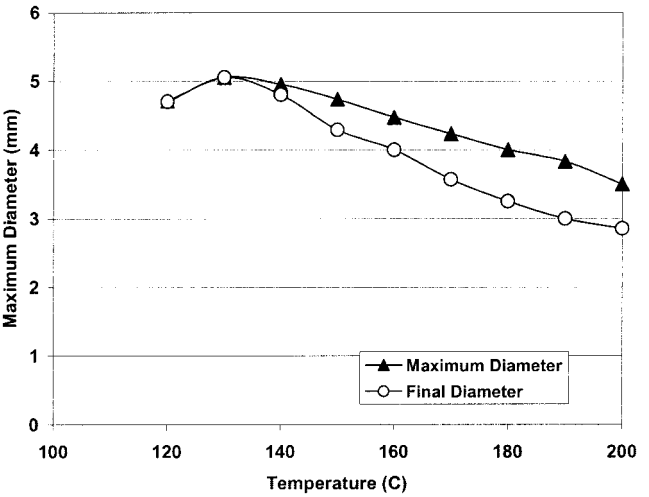


**Figure 8** Effect of processing temperature on the initial diameter.

## CONCLUSIONS

The fundamental mechanisms governing the volume expansion behavior of PP foams were determined on

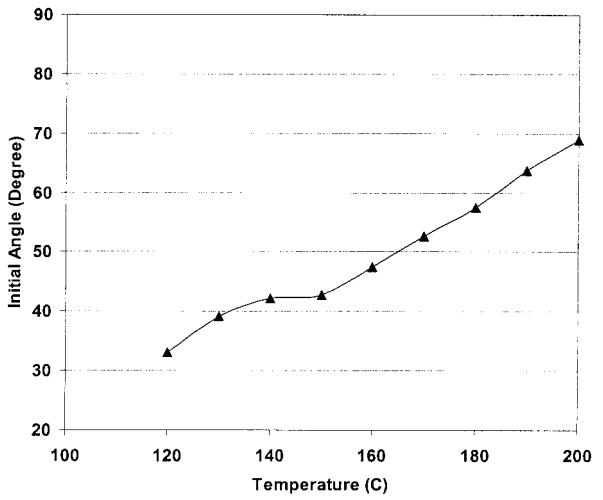


**Figure 7** Effect of processing temperature on the initial expansion.

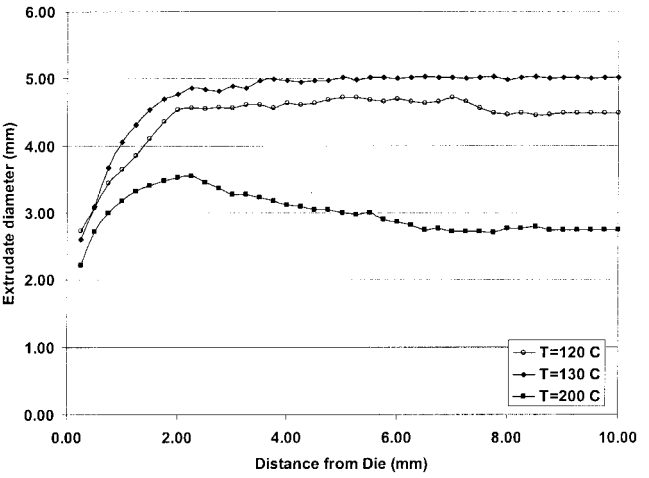


**Figure 9** Extrudate diameter as a function of the distance from the die.

the basis of experimental results. It turned out that either gas loss or polymer crystallization governed the expansion behavior of the PP foams. A progressive image scanning setup was configured to capture the images of the foamed extrudate coming out of the die on a PC. The images captured were analyzed, and the data were used to verify the proposed mechanisms. The processing window variation with the butane concentration, the change in the temperature ranges for the two governing mechanisms, and the sensitivity of melt temperature variations to the volume expansion ratio were discussed in detail on the basis of the obtained experimental results.

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