# A Study of Foam Extrusion Using a Chemical Blowing Agent

C. D. HAN, Y. W. KIM, and K. D. MALHOTRA, *Department of Chemical Engineering, Polytechnic Institute of New York, Brooklyn, N.Y. 11201*

## Synopsis

An experimental study of foam extrusion was carried out to determine the effect of processing variables on the quality of the foam produced. For the study, the chemical blowing agent azodicarbonamide was used, together with an activator and a nucleating agent, to produce thermoplastic foams of low-density polyethylenes. The quality of foam was determined from photomicrographs and the tensile properties of extrudate samples. It was found that the percent elongation correlates with the foam density and that the cell structure (cell size and its distribution) correlates with the ultimate tensile properties of the foam produced. Also, an experimental study was carried out to observe the growth of gas bubbles as the polymer melt containing a blowing agent flows through a rectangular channel constructed of quartz. This experiment helped us to interpret the *curved* pressure profile of the polymer melt containing a blowing agent as the melt approaches the die exit, whereas the polymer *without* a blowing agent shows a linear pressure profile.

## INTRODUCTION

In recent years, the polymer processing industry has become increasingly interested in the production of extruded foams[1-5] and injection-molded structural foams.[6-13] This is because, in many cases, lower density can be achieved, which makes the technique economically attractive. Also, the use of a blowing agent together with polymers can give rise to products (e.g., synthetic wood) which otherwise would be very difficult to obtain. For instance, the use of thermosetting acrylic monomers in plasticized PVC foams allows the compounder to produce foams having a wide spectrum of hardness and deformation resistance.

In producing plastic foams, two types of blowing agent are used, chemical and physical. Chemical blowing agents are commonly used to produce high-density foamed polyolefins. Among many such agents, azodicarbonamide has widely been used successfully in foam extrusion. They function by decomposing at and above a critical temperature and releasing a gas, for example, nitrogen, carbon monoxide, or carbon dioxide. The bubbles thus initiated may be dissolved into the hot melt under high pressure and will grow when the pressure is reduced as the molten polymer flows through an extrusion die.

Physical blowing agents are gases such as nitrogen, dichlorotetrafluoroethane, n-pentane, and others. They are introduced either as a component of

the polymer charge or under pressure into the molten polymer in the barrel of the extruder.

When many bubbles are initiated, their ultimate size will be small. If few bubbles are initiated, they will become quite large, which is generally not desirable. Therefore, in order to control the initial bubble size, nucleating agents (e.g., sodium bicarbonate) are often used. In addition, catalysts are also often used to promote the efficiency of the blowing agent.

In the foam process, using either chemical or physical blowing agents, one of the most important and yet difficult problems from the processing standpoint is to achieve intimate mixing of the blowing agent and nucleating agent in the polymer melt, and cooling the melt to the foaming temperature. There are a variety of practical techniques for achieving this, and they are the very carefully guarded secrets of the industries concerned. The monograph edited by Frisch and Saunders[14] describes various foam processing techniques.

In foam extrusion, as in conventional extrusion operations, the measurement of the rheological properties of the polymer melt, in this case containing dissolved gas, is of fundamental importance to the design of the processing equipment (e.g., screw and die designs). As one may surmise, the flow properties of a polymeric material in foam extrusion varies with the type of foaming agent, the amount of foaming agent, and also with the type and amount of other additives (e.g., plasticizers, modifiers, nucleators, etc.). Also important in foam extrusion is a better understanding of the mechanism of foam formation. In particular, the control of the nucleation and growth of gas bubbles is vitally important to the success of the process.

In this paper, we shall present our recent study of foam extrusion, which was concerned with achieving a uniform cell distribution and the desired physical properties of the foam produced by varying the processing conditions (e.g., temperature and pressure) and the geometry of the extrusion dies.

## EXPERIMENTAL

In the present study, we carried out three different types of experiment. They were (1) wall-normal stress measurement in a capillary die; (2) visual observation of the bubble growth phenomenon in a transparent test channel; (3) flat-film extrusion of thermoplastics using a chemical blowing agent.

### Wall-Normal Stress Measurement

The apparatus used in this experiment was essentially the same as described in earlier papers by Han,[15,16] except that a Static Mixer (Kenics Corp., Thermogenizer) was installed at the extruder outlet. The mixer was used in order to improve the dispersion of the blowing agent in the molten polymer. A schematic of the experimental apparatus is given in Figure 1a.

High-density polyethylene (Union Carbide, DMDJ 4306) was used as base polymer, and azodicarbonamide (National Polychemical, Kempore 125) was used as blowing agent. The polymer was mixed with the blowing agent by tumbling for about 2 hr, and the mixture was then extruded through a 1-in.-
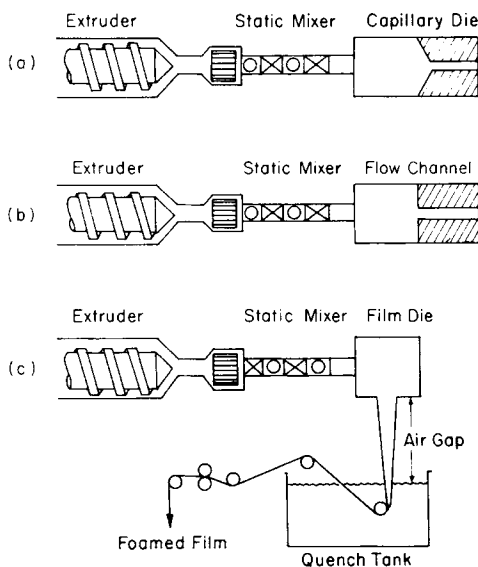
Fig. 1. Layout of experimental apparatus: (a) capillary extrusion apparatus; (b) flow visualization apparatus; (c) flat-film extrusion apparatus.

diameter Killion extruder equipped with variable-speed drive and on-off temperature controllers.

In the measurement of wall-normal stresses in a capillary die ($L/D$ = 4 and 20; $D$ = 0.125 in.), melt pressure transducers (Dynisco PT432) were used having a sensitivity of ±1% of the measured value. Details of the experimental procedures are described in earlier papers by Han.[15,16]

## Visual Observation of Bubble Growth in Extrusion

In this experiment, a rectangular flow channel constructed of quartz was used so that the growth of gas bubbles could be observed from the direction perpendicular to the direction of flow. The layout of the experimental setup is given in Figure 1b.

Motion pictures were taken of the bubbles flowing into and through the slit die section, using a special lens system to provide the proper magnification. The experiment was carried out under various extrusion conditions, i.e., for different values of temperature, flow rate, and die wall temperature.

## Flat-Film Extrusion Experiment

The layout of the experimental apparatus is given in Figure 1c. In this experiment, a flat-film die was used. The die was designed in such a way that the die land length and entrance angle were independently adjustable, as schematically shown in Figure 2. In this experiment, as the extrudate emerges from the die, it is cooled by passing it through a water tank. The air gap is the distance between the die exit and the surface of the water.

Low-density polyethylene (Union Carbide, DYNF 1) was used as base polymer, azodicarbonamide (National Polychemicals, Kenpore 125) as blowing agent, solid Ba–Cd (Cincinnati Milacron, Advastab ABC 50) as activator,

TABLE I
Experimental Polymer Formulations Used

| Material | Weight (by parts) |
|---|---|
| Formula #1 | |
| Low-density polyethylene | 100.0 |
| Azodicarbonamide | 0.5 |
| Formula #2 | |
| Low-density polyethylene | 100.0 |
| Azodicarbonamide | 0.5 |
| Talc | 0.2 |
| Formula #3 | |
| Low-density polyethylene | 100.0 |
| Azodicarbonamide | 0.5 |
| Talc | 0.2 |
| Calcium stearate | 0.2 |
| Formula #4 | |
| Low-density polyethylene | 100.0 |
| Azodicarbonamide | 0.5 |
| Solid Bd−Cd Activator | 0.5 |

and calcium stearate and magnesium silicate (talc) as nucleating agent. Specific formulations used in the present study are given in Table I.

The variables investigated were die entry angle (20° and 90°); die land length ($L/h$ ratio of 0 and 20; see Fig. 2); screw speed and die inlet pressure; die temperature; the effect of additives using formula #2 and #3; the effect of activator using formula #4; the effect of varying the air gap from 2 in. to 16 in.

Samples were collected and tested for the following: (a) foam quality by visual observation; (b) foam thickness and specific gravity; (c) tensile strength and per cent elongation; (d) foam structure using an optical microscope. The specific gravity was measured using a water displacement technique, and the tensile properties, using the Instron Tensile Testing machine.
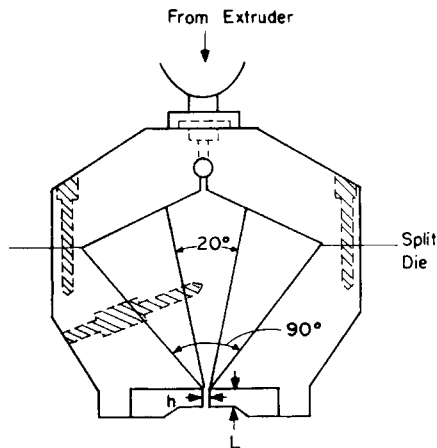


Fig. 2. Schematic of a film die used: $L$ is die land length and $h$ is die opening.

## RESULTS AND DISCUSSION

### Axial Pressure Profile and Bubble Growth in Extrusion Die

The rheological properties of a polymer containing blowing agents are of practical interest. The gas, which either evolved from the decomposition of a chemical blowing agent or was introduced as a physical blowing agent, is dissolved into the molten polymer at high pressure in the upstream of the extrusion die. However, as the melt approaches the die exit, the pressure of the flowing melt is reduced, giving rise to a rapid growth of bubbles, until the gas pressure in the bulk polymer is in equilibrium with that within the bubbles themselves.

It may be surmised, therefore, that the gas dissolved in the melt can affect the viscosity of the bulk polymer in the extrusion die. As a matter of fact, Blyler and Kwei[17] have reported that the melt viscosity of polyethylene was reduced by 20% when 0.5% by weight of a blowing agent was added. No systematic study has, however, been made on the effect of blowing agents (chemical or physical) on the pressure drops in the extrusion die, and hence on the viscosity of bulk polymers in foam extrusion.

Figure 3 gives axial profiles of wall-normal stress of high-density polyethyl-
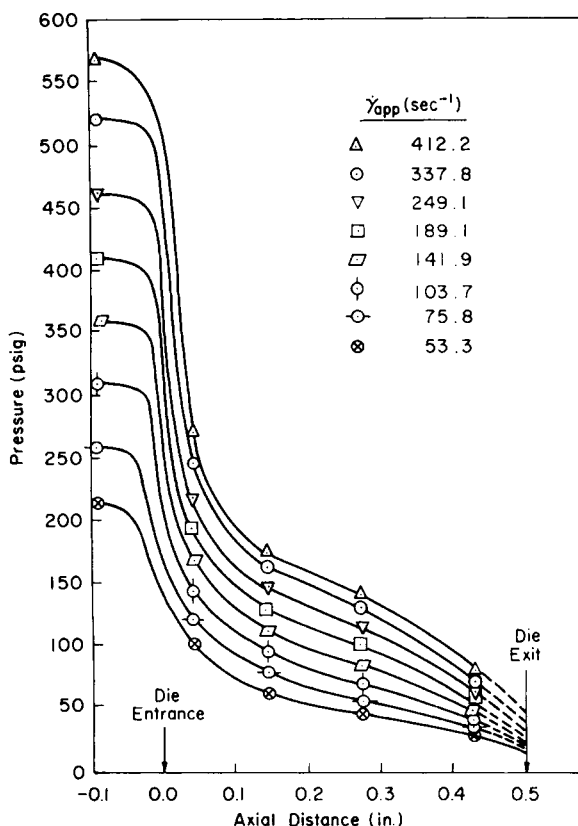


Fig. 3. Representative axial profiles for high-density polyethylene containing a blowing agent ($T = 200°C$).
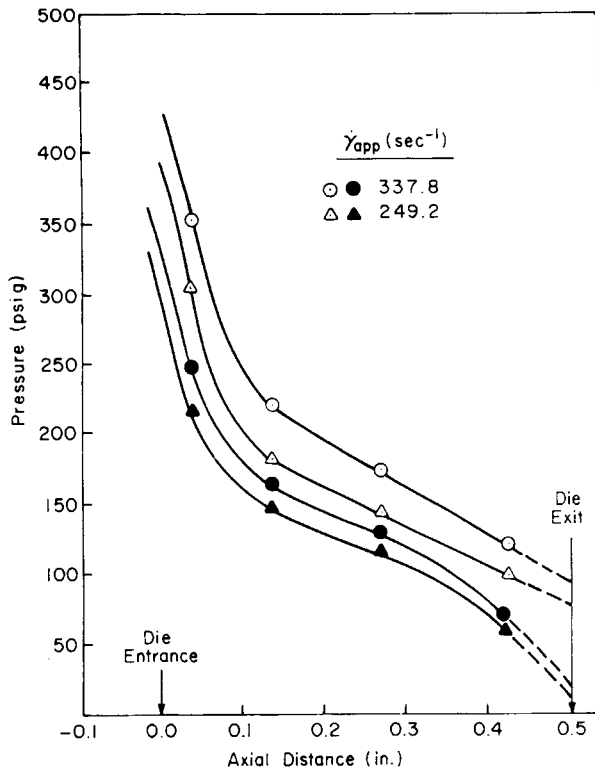
Fig. 4. Representative axial profiles for high-density polyethylene with and without blowing agent. Closed symbols are *with* blowing agent, and open symbols *without* blowing agent.

ene containing 0.3 wt-% of a blowing agent in a capillary having an $L/D$ ratio of 4. It is seen that the pressure profile starts to deviate from a straight line somewhere in the capillary as the melt approaches the die exit and that the pressure drop at the die entrance is exceedingly large. Figure 4 shows axial profiles of wall-normal stress of high-density polyethylene with and without a
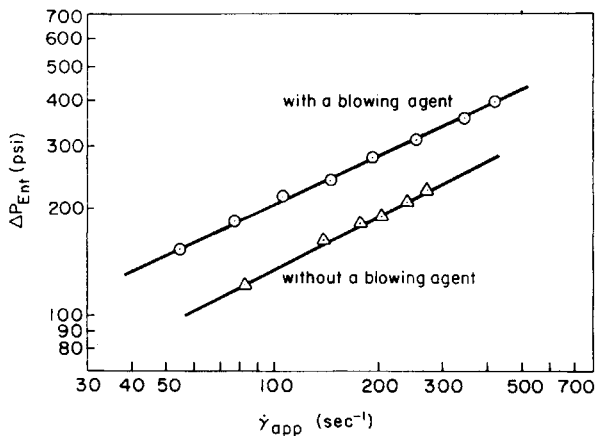


Fig. 5. Entrance pressure drop vs. apparent shear rate in the extrusion of high-density polyethylene with and without blowing agent ($T = 200°C$).

blowing agent (azodicarbonamide), measured along a capillary with an $L/D$ ratio of 4 ($D = 0.125$ in.). Two things can be seen in Figure 4. One is that, within the entire die length, the polymer with a blowing agent gives rise to lower wall-normal stresses than the polymer without a blowing agent. The other is that, after the melt passes the entrance region, the polymer with a blowing agent shows a curvature in its axial profile, whereas the polymer without a blowing agent shows a linear pressure profile.

That the pressure gradient is *not* constant in the capillary for the melt containing a blowing agent casts a doubt on the validity of determining the rheological properties from the wall normal stress measurements. In other words, the wall shear stress $\tau_w$ to be calculated from the expression

$$\tau_w = \left(-\frac{\partial p}{\partial z}\right)\frac{D}{4} \tag{1}$$

is not constant along the capillary for the melt containing blowing agent. In eq. (1), $(-\partial p/\partial z)$ is the pressure gradient at the position $z$, and $D$ is the capillary diameter.

Another interesting observatin that one can make in Figure 3 is the entrance pressure drop that occurs in the flow of polymer melt containing a blowing agent. Figure 5 shows plots of entrance pressure drop versus apparent shear rate for high-density polyethylene melts with and without a blowing agent. It is seen that the melt containing a blowing agent gives rise to much greater entrance pressure drops than the melt containing no blowing agent. The difference in the entrance pressure drop between the two curves in Figure 5 may be attributable to a possible evolution of the dissolved gas from the molten polymer in the form of small bubbles (i.e., as nuclei), as the melt passes through the die entrance.

Figure 6 gives pictures of the bubble growth phenomenon when a molten polyethylene containing a blowing agent is extruded through a transparent rectangular channel (see Fig. 1b) at different flow rates. It is seen that: (a) at low flow rates (see Fig. 6a) bubbles start to grow right after the die entrance, giving rise to a large number of big bubbles near the die exit; (b) at medium flow rates (see Fig. 6b), bubbles start to grow somewhere in the middle of the slit section; (c) at high flow rates (see Fig. 6c), the location where bubbles start to grow moves toward the die exit. In other words, as the flow rate is increased, the chances of bubble growth in the die become less. It should be pointed.out that an increase in flow rate accompanies an increase in pressure.

From the experimental observations given in Figure 6, it may be concluded that the curved pressure profiles given in Figures 3 and 4 for the melt containing a blowing agent may be attributable to the rapid growth of bubbles as the melt approaches the die exit. In other words, there would be a critical value of pressure below which small bubbles start to grow. Of course, the critical pressure depends on the radius $R$ of the initial bubble, interfacial tension $\sigma$, and the pressure difference $\Delta p$ between the surrounding fluid (i.e., polymer melt in the present case) and inside the bubble. Thus, at equilibrium, the following force balance should hold[18]:

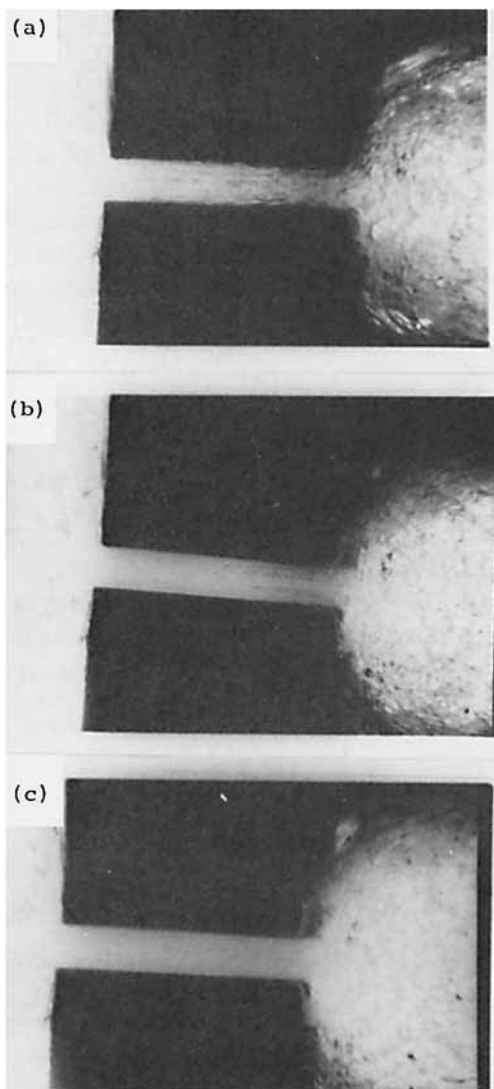$$\Delta p = \frac{2\sigma}{R} \tag{2}$$

Fig. 6. Representative photographs showing bubble growth phenomenon in the flow of a molten polyethylene, containing blowing agent (azodicarbonamide), through a rectangular channel at differeent flow rates: (a) $Q = 4.21$ cc/min; (b) $Q = 9.25$ cc/min; (c)$Q = 15.01$ cc/min.

Note that the interfacial tension $\sigma$ may vary with the type of polymer used, and the radius $R$ of the initial bubble depends on the type of blowing agent used as well as the additives (e.g., nucleating agent and activator).

To summarize, it has been shown above that the pressure profiles of a polymer melt containing a blowing agent in an extrusion die depend on the state of the bubbles dispersed in the melt. It should be mentioned that control of the bubble size and its distribution is vitally important to produce foams of uniform quality. During the extrusion operation, the size of the foam cell (i.e., bubble) changes with time because of diffusion of gas from the liquid phase into the cell and also by possible coalescence of gas bubbles. Hence,

the stabilization of the foam cell is important. A better understanding of the mechanism of nucleation and growth of bubbles, and of the coalescence of bubbles, in a foaming process is very important to obtaining a product of desired quality.

### Effect of Processing Variables on Foam Quality and Tensile Properties

Table II shows the effect of the die entry angle on foam quality. The thickness of the foam sheet produced is less for the smaller angle and the specific gravity is greater. Foam quality, as judged by appearance, is found to be better at the smaller angle. This may be due to the increased entrance pressure drops so obtained.

In order to determine whether the die land length would affect the foam quality, two extremes were used in our experiment. They were the die with zero land length, wherein the material exits immediately after the converging section, and the die fitted with a land length $L$ of 0.4-in. Since the die opening $h$ is 0.020 in., this would mean an $L/h$ ratio of 20. Table III shows that the die length appears to have very little effect on the foam quality.

TABLE II
Variation of Foam Quality with Die Entry Angle[a]

| Die entry angle, degrees | Foam thickness, mils | Specific gravity | Tensile Strength, psi | Percent elongation, % | Foam quality |
|---|---|---|---|---|---|
| 90 | 15.8 | 0.530 | 760 | 185 | fair, elongated bubbles present due to coalescence of bubbles and subsequent stretching |
| 20 | 8.5 | 0.625 | 1040 | 240 | better foam, fewer and smaller bubbles; more uniform appearance |

[a] Operating conditions: screw temperature, 200°C; die temperature, 200°C; screw speed, 30 rpm; land length $(L/h)$, 20; formula, #1; haul-off speed, 5.65 ft per min; air gap, 5 in.

TABLE III
Variation of Foam Quality with Die Land Length[a]

| Land length $(L/h)$ | Foam thickness, mils | Specific gravity | Tensile Strength, psi | Percent elongation, % | Foam quality |
|---|---|---|---|---|---|
| 0 | 19.8 | 0.555 | 605 | 136 | fair; nonuniform bubble size distribution; bubbles elongated due to stretching |
| 20 | 23.5 | 0.530 | 545 | 144 | same as above |

[a] Operating conditions: screw temperature, 220°C; die temperature, 220°C; screw speed, 70 rpm; die entry angle, 20°; formula, #1; haul-off speed, 5.65 ft per min; air gap, 5 in.

TABLE IV
Variation of Foam Quality with Screw Speed[a]

| Screw rpm | Die inlet pressure, psi | Foam thickness, mils | Specific gravity | Percent elongation, % | Tensile strength, psi | Foam quality |
|---|---|---|---|---|---|---|
| 50 | 150 | 16.9 | 0.590 | 190 | 620 | fair; some non-uniformity in bubble sizes; bubbles are elongated; overall thickness is uniform |
| 70 | 220 | 21.2 | 0.590 | 120 | 600 | rougher surface, non-uniformity in overall thickness due to surging at the extruder exit |
| 95 | 270 | 28.0 | 0.570 | 135 | 600 | same as above, but ripple marks caused by surging clearly visible |

[a] Operating conditions: screw temperature, 200°C; die temperature, 200°C; die entry angle, 20°; land length ($L/h$): 20; take-up speed, 5.65 ft per min; formula, #1; air gap, 2 in.

Extruder screw speeds were increased from 50 rpm to 95 rpm, and Table IV shows how this affected the foam quality. From Table IV, one can see that the quality of foam deteriorates above 50 rpm screw speed. This implies that flow instabilities set in at higher shear rates. The die inlet pressure (i.e., the pressure in the reservoir) was measured by a pressure gauge located just after the adaptor. As would be expected, the melt pressure increased with increasing screw speed. Based on this observation, it can be said that the effect of die land length on foam quality has not been fully established because the experiment was run under screw speed conditions (>50 rpm) which produces poor quality foam.

TABLE V
Variation of Foam Quality with Melt Temperature[a]

| Die temp., °C | Die inlet pressure, psi | Foam thickness, mils | Specific gravity | Tensile strength, psi | Percent elongation, % | Foam quality |
|---|---|---|---|---|---|---|
| 200 | 220 | 21.2 | 0.590 | 600 | 120 | rough surface; non-uniformity in thickness due to surging at the extruder exit |
| 160 | 325 | 23.0 | 0.615 | 800 | 150 | surface still rough, but thickness is more uniform |

[a] Operating conditions: screw temperature, 200°C; die entry angle, 20°; land length ($L/h$), 20; screw speed, 70 rpm; take-up speed, 5.65 ft per min; formula, #1; air gap, 2 in.

TABLE VI
Variation of Foam Quality with Air Gap Distance[a]

| Air gap, in. | Foam thick-ness, mils | Specific gravity | Tensile strength, psi | Percent elonga-tion, % | Foam quality |
|---|---|---|---|---|---|
| 2 | 22 | 0.530 | 765 | 128 | fair; large elongated bubbles visible |
| 6 | 33 | 0.545 | 820 | 175 | good; bubble distribu-tion is more uniform |
| 8 | 42 | 0.555 | 820 | 180 | same as above |
| 16 | 56.8 | 0.530 | 735 | 208 | same as above |

[a] Operating conditions: screw temperature, 180°C; die temperature, 180°C; die entry angle, 20°; die land ($L/h$), 20; screw speed, 70 rpm; take-up speed, 5.65 ft per min; formula, #4.

Chemically blown foam is very sensitive to the temperature of the polymer melt. From Table IV, one sees that the surface of the foamed extrudate was rough. In order to obtain a smoother surface and more uniform foam, the temperature of the die was reduced from 200° to 160°C. The effect of this change is shown in Table V. Even though a smooth skin was not formed at the lower temperature, foam uniformity did improve. Since the die tempera-ture was separately controlled, via a solid-state thermistor control, it was quite easy to change it.

Figure 7 shows the relationship of ultimate tensile strength to specific grav-ity of the foam. Figure 8 is an attempt to show that the quality of foam, as judged by appearance, can be quantitatively determined by measuring per cent elongation.

Interpreting the above, one can say that the tensile strength increases with specific gravity and is not much dependent on cell size or cell distribution, whereas the percent elongation is strongly dependent on the foam quality.
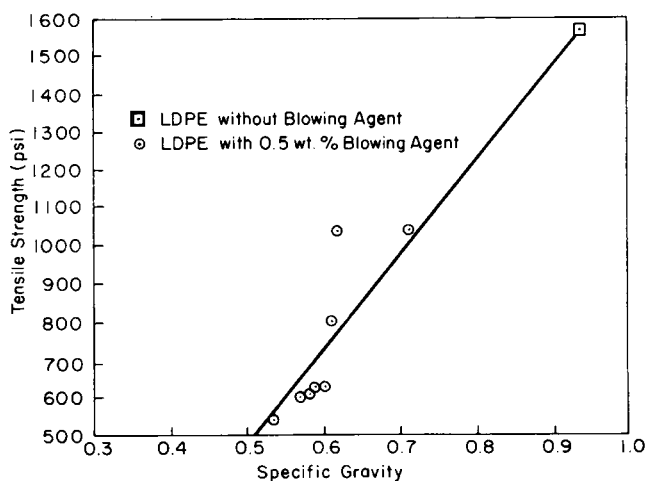


Fig. 7. Tensile strength vs. specific gravity for low-density polyethylene foam with a blowing agent (azodicarbonamide).
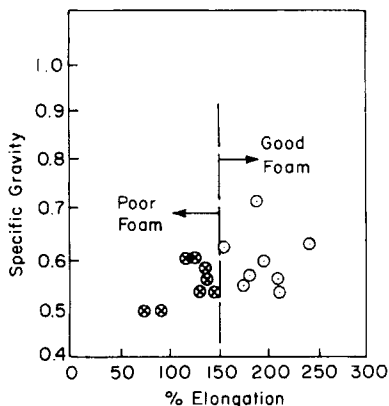
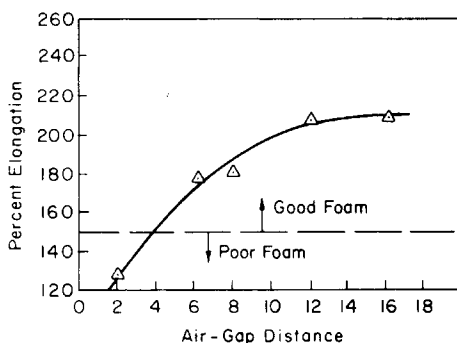Fig. 8. Specific gravity vs. percent elongation for low-density polyethylene foam.

Fig. 9. Percent elongation vs. air gap distance for low-density polyethylene foam.

Table VI shows how the foam quality generally improves as the air gap is increased in steps from 2 to 16 in. A further lowering of the water level resulted in inadequate cooling. It is further seen in Table VI that, as the air gap is increased, the following happens: (1) the thickness of the foam film increases but the width decreases; (2) the specific gravity remains unaffected; (3) the tensile strength remains fairly constant; (4) there is a dramatic increase in elongation with the increase in air gap from 2 to 6 in. On further increasing the air gap, the elongation increases slowly and then levels off. This is shown in Figure 9.

It has been shown in a previous study[19] that the void morphology (hence, foam quality) is affected by the crystallinity of the polymer. It appears, then, that further research is needed to quantitatively relate the effect of the degree of the crystallinity of the polymer to foam quality and bubble distribution.

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