

A Study of Sandwich Foam Coextrusion

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Synopsis

An experimental study of sandwich foam coextrusion was carried out, using a sheet-forming die with a feedblock. Polymers used for the experiment were low-density polyethylene (LDPE) for the outer layers and ethylene-vinyl acetate (EVA), with the chemical blowing agent azodicarbonyl diamide, for the foamed core component. The present study has shown that the cell size and its distribution in the foamed core and the mechanical properties of the sandwiched foam product can be controlled by a judicious choice of the thickness ratio of the core to skin components, the melt-extrusion temperature, and the concentration of chemical blowing agent.

INTRODUCTION

The "sandwich foam coextrusion process" is a relatively new polymer processing technique that combines the film coextrusion process and the foam extrusion process. In producing sandwiched foam products, the core-forming polymer B containing a blowing agent is coextruded with the skin-forming polymer A, as shown schematically in Figure 1. A large number of combinations of polymer systems may be used for the skin and core components of a sandwiched foam. In the selection of materials, both the core-forming polymer B and the skin-forming polymer A can be the same (except that B contains a blowing agent), or they can be different polymers. If desired, A may contain an additive (or additives) such as an antistatic or a flame retardant agent.

A sandwiched foam product may also be obtained by means of the injection molding process. The so-called "sandwich molding process" was first reported by Oxley and Sandiford.¹ According to them, two polymers are injected into the mold through a common sprue with the following sequence: (a) The skin-forming polymer A is injected to partially fill the mold. (b) The core-forming polymer B containing a blowing agent is injected into the mold, forcing the skin-forming polymer A outward. In this step, the core-forming polymer B fills the mold completely and ensures a good surface finish with a uniform skin layer of polymer A. (c) For the second time, the skin-forming polymer A is injected into the mold cavity to clear the foamable polymer B in the sprue. This step is to prevent the residual foamable polymer B remaining in the sprue from contaminating the skin-forming polymer A of the next molding. (d) The mold is opened a small distance so that the core-forming polymer B can now foam to give a uniform foam structure within a thin skin of polymer A that contains *no* foam residue.

It is of interest to note that in the use of either the sandwich coextrusion process or the sandwich molding process, the core-forming polymer need not contain a blowing agent. For instance, the sandwich coextrusion process has been used to produce multilayer flat films,^{2,3} and the sandwich molding process to produce molded articles with an enhanced reinforcement in the skin.¹ As may be sur-

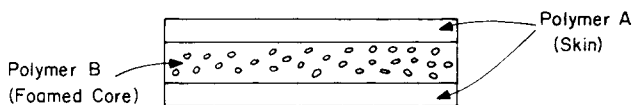


Fig. 1. Schematic of the cross section of a sandwiched foam product.

mised, the flow behavior in sandwich molding is far more difficult to control than that in sandwich coextrusion. In their recent paper, White and Lee⁴ have reported the flow behavior of two incompatible polymers being sandwich-molded into a rectangular mold cavity.

In the present paper, we report the results of our recent experimental study of sandwich foam coextrusion.

EXPERIMENTAL

The apparatus used in the present study consists of a die, two extruders, a Static Mixer, a quench tank, and a take-up device. Figure 2 gives a schematic of the experimental apparatus. The die used for the sandwich foam coextrusion has a feedblock and a slit section. The slit section is 4.0 in. in width, 2.5 in. in length, and 0.04 in. in thickness (or die opening). The feedblock consists of three sections, each capable of receiving a separate feed stream. A photograph of the die used is given in Figure 3.

The die is provided with three melt-pressure transducers (Dynisco PT 422) in the axial direction (along the centerline of the slit channel) at distances of 0.5, 1.0, and 1.5 in. from the die exit. The pressure measurements were used to determine the axial pressure gradient of the fluids under test. The die is heated electrically with band heaters, and temperature is controlled by thermistor-operated thermal regulators.

In the present study, the two outer feedports were used to receive the skin-forming polymer and the center feedport, to receive the core-forming polymer containing a blowing agent. The chemical blowing agent was first blended with the core-forming polymer by means of a tumbling operation, and the mixture was fed to an extruder that is connected to a commercially available Static Mixer (Kenics Corp.). The skin-forming polymer is fed to an extruder and to the feedblock (see Fig. 2). Polymer A forms the skin and polymer B, the foamed core, as shown schematically in Figure 1. The thickness of the skin layer is controlled by regulating the ratio of the flow rate of polymer A to that of polymer B.

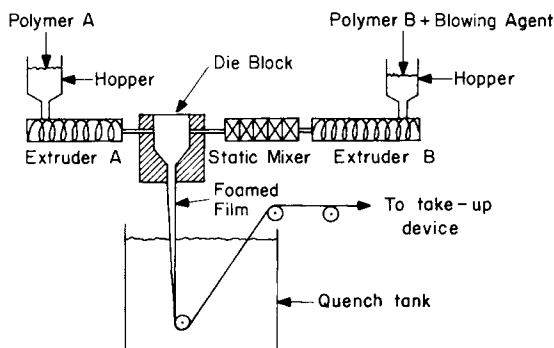


Fig. 2. Schematic of the layout of the apparatus.

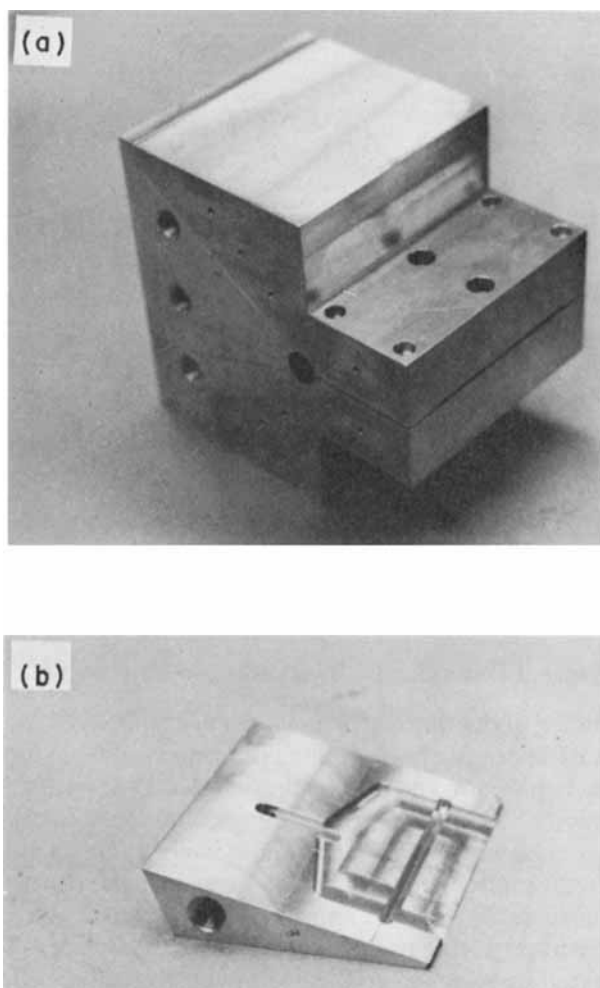


Fig. 3. Photograph of the die used.

The polymers used were low-density polyethylene (LDPE) (Union Carbide, DYNF-1) and ethylene-vinyl acetate (EVA) (Union Carbide, DQDD 1868). Figures 4 and 5 give plots of melt viscosity versus shear rate for LDPE and EVA, respectively. In all the runs, the EVA was used as the core component containing a blowing agent, sandwiched between two layers of LDPE. That is, LDPE/EVA/LDPE was the sandwich foamed in the present study. As blowing agent, azodicarbonamide (National Polychemicals, Kempore 125) was used.

In the extrusion experiment, after the flow was equilibrated for a given setting of flow rate, measurements were taken of wall normal stresses of the molten polymers with the aid of the melt pressure transducers. Details of the experimental procedure for the pressure measurement and its interpretation are given elsewhere.⁵⁻⁷

The processing variables investigated were the extrusion temperature, volumetric flow rate, and the concentration of the blowing agent in the foamed core.

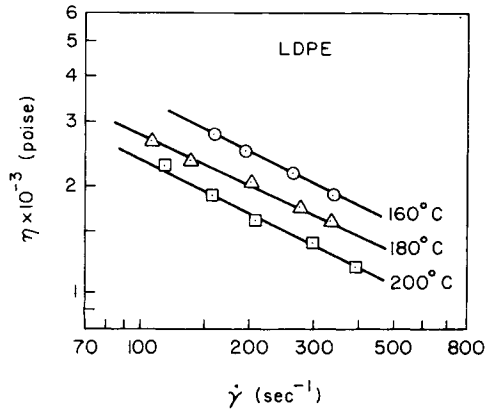


Fig. 4. Melt viscosity vs shear rate for low-density polyethylene.

Extrudate samples were collected and used later to examine the foam structure and to determine the mechanical properties.

RESULTS AND DISCUSSION

Rheological Behavior of Gas-Charged Molten Polymers

An understanding of the rheological behavior of gas-charged molten polymers is of fundamental importance to controlling the structural foam process. As may be surmised, the bubble size and its distribution during processing influence the rheological behavior of a gas-charged molten polymer. It is a well-established fact that the gas evolved from the decomposition of a chemical 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. This is recently been demonstrated by Han and coworkers^{8,9} who photographed the motion of gas bubbles growing in the gas-charged molten polymer flowing through a slit die with glass windows.

Figure 6 gives typical axial pressure profiles at 200°C of the LDPE/EVA-

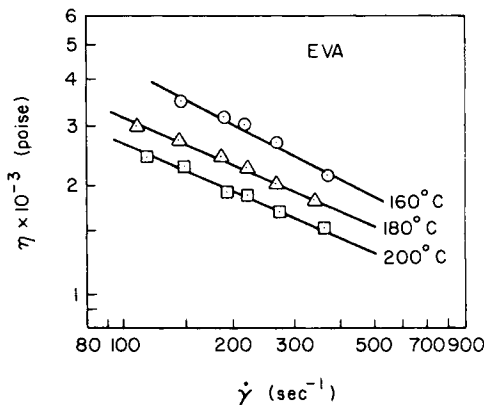


Fig. 5. Melt viscosity vs shear rate for ethylene-vinyl acetate.

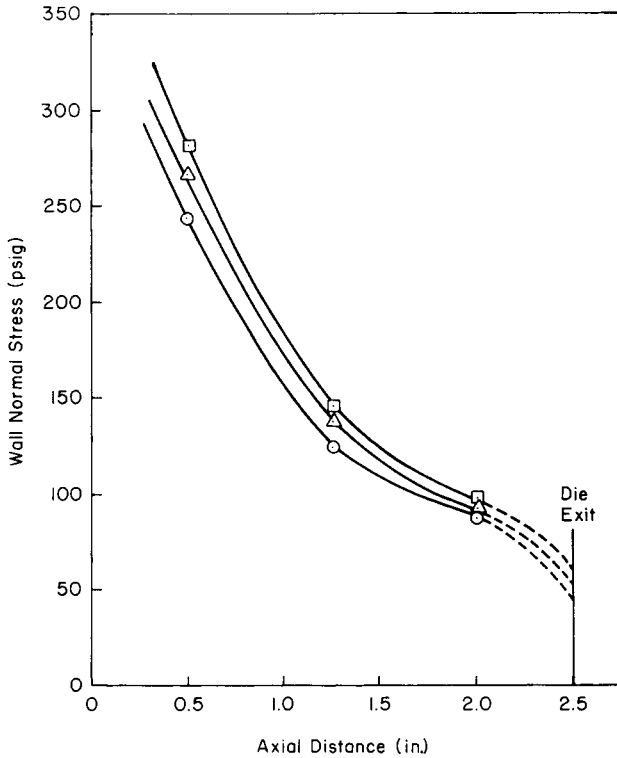


Fig. 6. Axial pressure profiles for LDPE/EVA(foam)/LDPE system. Q_{LDPE} (g/min): (□) 33.12; (Δ) 23.47; (○) 12.24. Q_{EVA} (g/min): (□) 21.56; (Δ) 21.56; (○) 21.56. $T = 200^{\circ}\text{C}$; 0.3% blowing agent.

(foam)/LDPE system containing 0.30 wt-% of blowing agent in the EVA. Similar plots at 240°C are given in Figure 7 for the system containing 0.6 wt-% of blowing agent. It is seen in Figures 6 and 7 that pressure (more precisely stated, wall normal stress) increases with flow rate and decreases with extrusion temperature. Note in Figures 6 and 7 that pressure gradients deviate from straight lines downstream in the die as the gas-charged polymer approaches the die exit. This observation is consistent with an earlier observation.⁸ It should be pointed out that molten polymers *without* a blowing agent yield constant pressure gradients downstream in the die.⁵⁻⁷

That the pressure gradient (i.e., the slope of the pressure profile) varies as the gas-charged molten polymer approaches the die exit indicates that the flow property varies in that region of the die. Note that when the pressure gradient ($-\partial p/\partial z$) is constant, the wall shear stress τ_w is determined⁷ by

$$\tau_w = \left(\frac{-\partial p}{\partial z} \right) \frac{h}{2} \quad (1)$$

where h is the opening of the slit die.

Note further that a gas bubble will start to grow when the external pressure is lower than the pressure inside the bubble such that, at equilibrium, the following force balance should hold:

$$\Delta p = 2\sigma/R \quad (2)$$

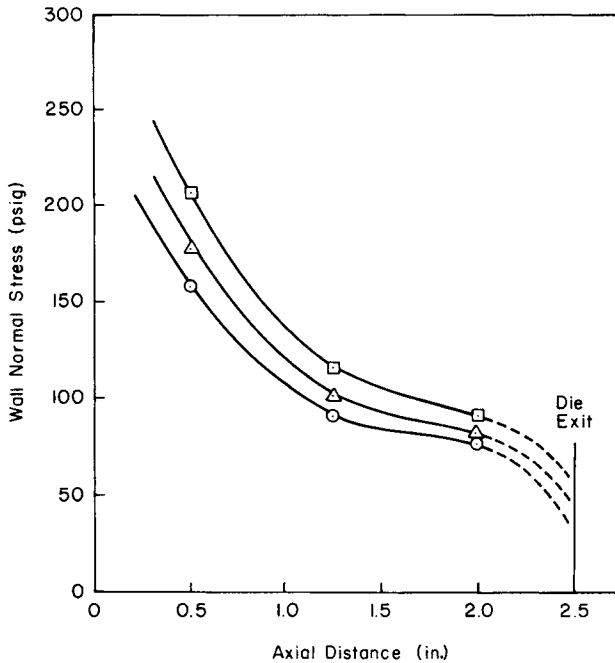


Fig. 7. Axial pressure profiles for LDPE/EVA(foam)/LDPE system. Q_{LDPE} (g/min): (◻) 39.08; (Δ) 23.18; (○) 13.26. Q_{EVA} (g/min): (◻) 25.31; (Δ) 25.31; (○) 25.31. $T = 240^{\circ}\text{C}$; 0.6% blowing agent.

in which Δp is the pressure difference between the surrounding fluid (i.e., polymer melt in the present case) and inside the bubble, σ is the interfacial tension, and R is the radius of the initial bubble. It is therefore important to maintain the external pressure greater than the pressure inside the bubble if one wishes to suppress bubble growth in the extrusion die. In other words, a critical value of pressure would exist below which small bubbles start to grow. According to eq. (2), the critical pressure depends on the radius R of the initial bubble.

It is therefore important to recognize that in foam extrusion, once bubbles start to grow somewhere in the die, the density of the fluid containing bubbles changes in that region of the die, and therefore shear rate is very difficult to define. Under such conditions, the determination of fluid viscosity is not possible unless one has information on the fluid density at various positions along the direction of flow. Unfortunately, however, relatively little has been reported in the literature^{8,10,11} which deals with the flow behavior of gas-charged molten polymers, whereas during the past decades extensive studies have been reported in the literature dealing with the rheological determination of homopolymers, polymer blends, and filled polymers in the molten state, as summarized in the recent monograph of Han.⁷ As pointed out by Throne,¹² the density of the gas-charged molten polymer depends on the solubility of gas in the melt, which in turn depends on the pressure profile along the die axis. In other words, bubble growth in foam extrusion depends on the pressure profile in the die.

It can be concluded that a better understanding of the phenomenon of bubble growth in a viscoelastic molten polymer is of fundamental and practical importance in controlling the rheological properties of gas-charged molten polymers. An investigation of this problem will be reported in a forthcoming publication.⁹

Effect of Temperature, Thickness (Skin/Core) and Concentration of Blowing Agent on Cell Size and Its Distribution

Some representative photographs displaying the foam morphology are given in Figures 8–10. The extrusion temperatures, the concentration of blowing agent, and the thickness ratio of skin to core components used in obtaining the extrudate samples shown in Figures 8–10 are given in Table I.

Photographs of the sandwich foam sample were taken with the help of an SLR camera (35 mm) fitted with close-up lenses. The photographs are of the composite foam sample taken in the longitudinal direction (i.e., from the top if the sandwich foam sample is placed on a flat surface), and the magnifications of the photographs were maintained constant at a value of 4. The bubble diameters in the sandwich foam samples were in the range of 0.25–3 mm.

Comparing Figures 8(a) and 9(a) and Figures 8(b) and 9(b), it is seen, as expected, that the number of nucleation sites is increased with an increase in concentration of the blowing agent at the same processing temperature. Further, the cell size at a lower concentration of blowing agent is smaller than the cell size

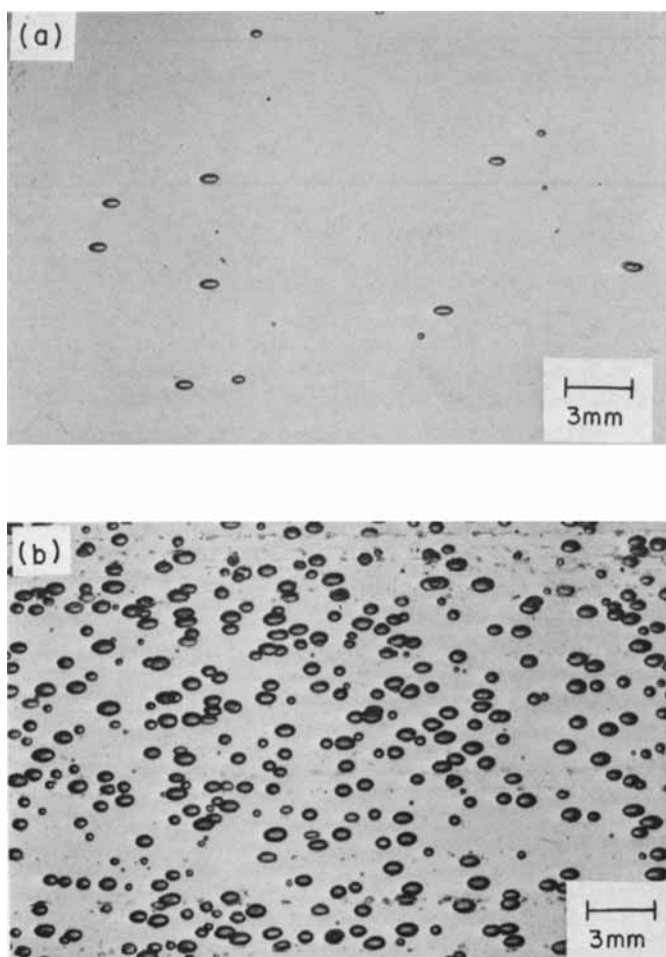


Fig. 8. Foam morphology of the samples extruded at 200°C containing 0.3 wt-% of blowing agent.

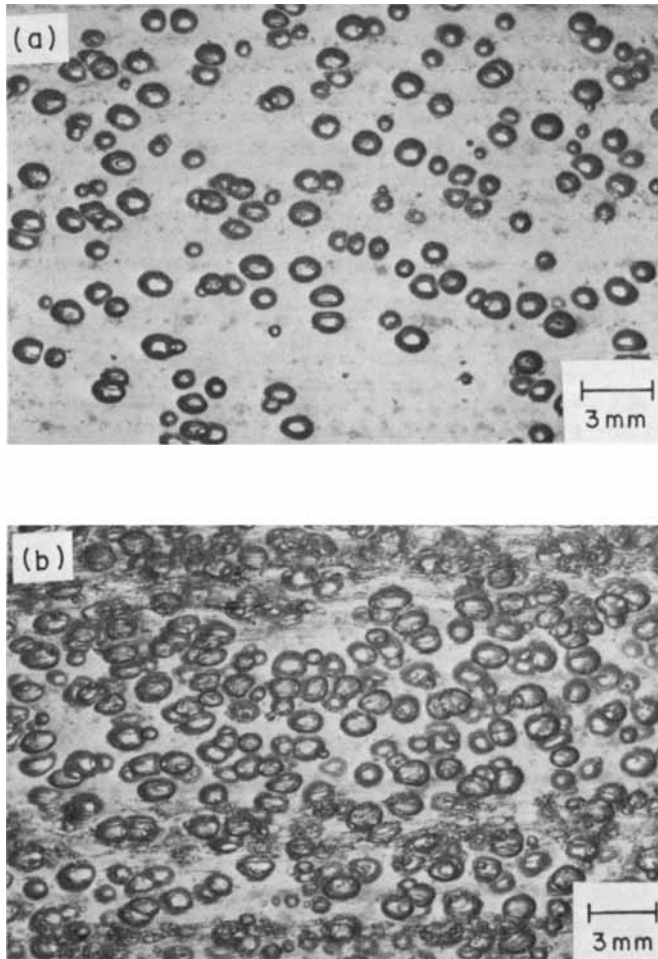


Fig. 9. Foam morphology of the samples extruded at 200°C containing 0.6 wt-% of blowing agent.

at a higher concentration of blowing agent. At a lower concentration of blowing agent, the number of nucleation sites is less, and hence the total void volume is less, resulting in an increase in melt viscosities. This effectively leads to a suppression of bubble growth and hence a reduction in cell size. On the other hand,

TABLE I
Processing Conditions Describing the Foam Morphology Given in Figures 8 to 10

Sample identification	Extrusion temperature, °C	Concentration of blowing agent, wt-%	Thickness ratio (skin/core)
8a	200	0.3	0.61
8b	200	0.3	0.18
9a	200	0.6	0.55
9b	200	0.6	0.16
10a	240	0.6	0.50
10b	240	0.6	0.18

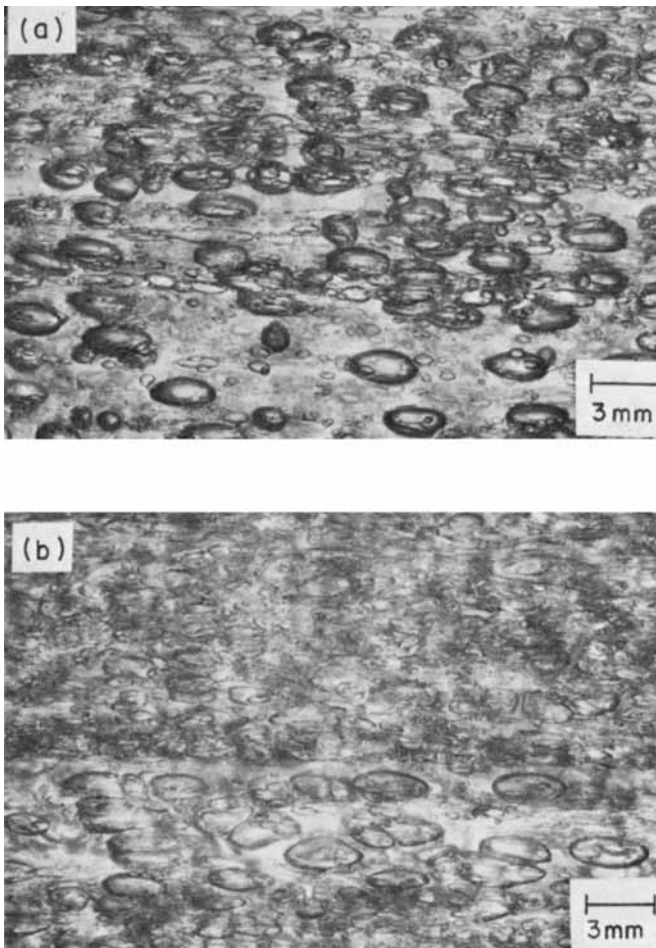


Fig. 10. Foam morphology of the samples extruded at 240°C containing 0.6 wt-% of blowing agent.

at higher concentrations of blowing agent, we have a decrease in melt viscosities, resulting in increased bubble growth and cell size.

Comparing Figures 9(a) and 10(a) and Figures 9(b) and 10(b), it is seen that the number of nucleation sites is approximately the same, but there is an increase in cell size as the melt extrusion temperature is increased. At higher temperatures, viscosities of the components used in processing are reduced (see Figs. 4 and 5). This allows for an increase in bubble growth and hence an increase in cell size. At lower temperatures, the viscosities of the components increase, resulting in a decrease in cell size.

Comparing Figures 8(a) and 8(b), Figures 9(a) and 9(b), and Figures 10(a) and 10(b), it is seen that there is an increase in cell size as the skin-to-core ratio is decreased (see Table I for the thickness ratio). This is because, as the skin-to-core ratio is decreased, the wall pressure and pressure gradient (hence wall shear stress) in the die are reduced. This facilitates bubble growth and results in bigger cell sizes. On the other hand, at higher skin-to-core ratios, the wall pressure and pressure gradient in the die are increased. This effectively suppresses bubble growth and results in a reduction of cell size.

From the above discussion, the following conclusions can be drawn: (1) An increase in the concentration of blowing agent results in an increase in the number of nucleation sites and cell size. (2) An increase in the processing temperature results in an increase in cell size. (3) An increase in the skin-to-core ratio results in a decrease in cell size in the foamed core.

Effect of Temperature, Skin-to-Core Ratio, and Concentration of Blowing Agent on Tensile Modulus

Figure 11 shows the relationship between the tensile modulus and the skin-to-core ratio at various temperatures for the LDPE/EVA(foam)/LDPE system. The tensile modulus experiments were carried out on an Instron testing machine. The specimens of the sandwich foam sample used in the determination of the tensile modulus were 4.0 in. in length and 0.5 in. in width. The tensile modulus measurements were carried out in the machine direction for the sandwich foam samples. The Instron cross-head speed was maintained constant at 2 in./min, and the test was run at a constant span of 2.0 in. During the testing it was observed that when the skin was very thick and the bubble size and bubble density was very small, we observed necking, very similar to the necking observed while testing unfoamed low-density polyethylene films, and the failure observed in the sandwich foam specimen was very similar to that observed during the testing of unfoamed low-density polyethylene samples. However, when the skin was very thin and the bubble size and foam density were much greater, there were "weak spots" in the specimen due to the larger bubble sizes and, in some cases, due to the coalescing of these bubbles. When these specimens were stretched, necking might have been initiated as in the unfoamed low-density polyethylene

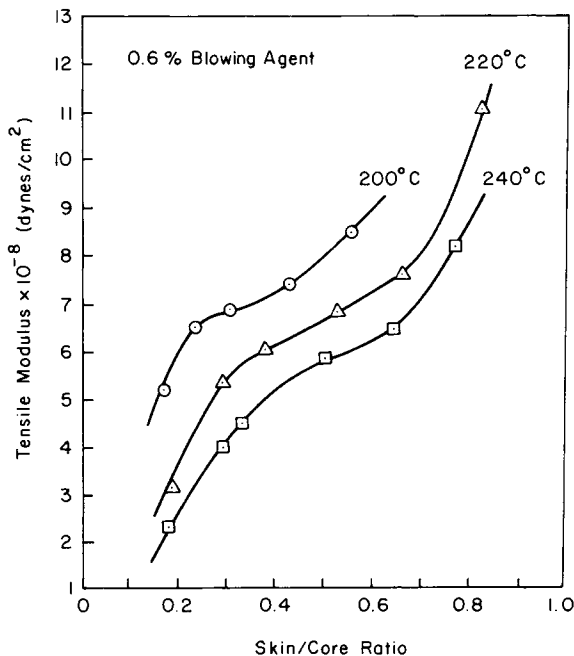


Fig. 11. Tensile modulus vs thickness ratio of the skin to core components for the samples extruded at different temperatures.

specimens; but at very low elongations, failure suddenly occurred at the "weak spots" in the specimen.

The tensile modulus of the sandwich foam sample is found to increase as the thickness ratio of the skin to core component is increased. This can be explained by the fact that, as the skin-to-core ratio is increased, the pressure and pressure gradient (hence wall shear stress) in the die are increased. This suppresses the growth of bubbles and results in smaller bubble sizes. On the other hand, as the skin-to-core ratio is decreased, the wall shear stresses in the die are decreased. This allows for greater bubble growth, resulting in an increase in the size of bubbles. In extreme cases, where the skin is very thin, the bubbles may start to coalesce and may even rupture the skin (see, for instance, Fig. 10). Effectively, this increases the total volume of the voids in the foam sample, which causes a decrease in its tensile modulus.

It is also seen in Figure 11 that the tensile modulus of the sandwich foam samples decreases as the processing temperature is increased. Note that at higher temperatures there is a reduction in the melt viscosities of the components being processed (see Figs. 4 and 5). This allows for an increase in bubble growth and hence an increase in the size of the bubbles. The total volume of voids in the sandwiched foam is thus increased, resulting in a decrease of the tensile modulus. On the other hand, when the processing temperature is decreased, there is an increase in the melt viscosities of the components being processed. This results in a suppression of bubble growth and leads to a decrease in the size of the bubbles. The total void volume having thus been reduced, the tensile modulus of the foamed sample increases.

In the past, other researchers also investigated mechanical properties of thermoplastic foams. Hobbs¹³ has experimentally determined the load-deflection characteristics of structural foams of Lexan and Noryl resins, and these results were compared with those calculated from several idealized beam models as well as from a stress function analysis. In his theoretical study, Hobbs has considered the structural foam as a two-component beam having a skin modulus and a core modulus and has put forth a linear and a nonlinear model to describe the specimen. Mehta and Colombo¹⁴ have determined the mechanical properties of polystyrene foams and have derived an empirical formula to relate the foam modulus with foam and resin properties, e.g., modulus of the solid polymer, foam density, solid polymer density, and structure factor (which is a function of the ratio of the cell dimension in the machine direction to the cell dimension in the transverse direction).

From the above discussion we can conclude that (1) an increase in processing temperature effectively causes a decrease in the tensile modulus and (2) an increase in thickness ratio (skin/core) causes an increase in the tensile modulus. It has been observed also that an increase in the concentration of blowing agent results in a decrease of the tensile modulus. This is as expected because the total void volume is increased with the amount of blowing agent used.

CONCLUSIONS

In sandwich foam coextrusion, one can suppress the bubble growth very effectively by using a very viscous polymer as the skin-forming component. This is because the more viscous the skin-forming polymer, the higher the pressure

(and pressure gradient) will be. From the point of view of practical use, a sandwiched foam product having protective layers of hard skin (e.g., fiber glass-filled resin) is of commercial importance. In view of the fact that the use of a viscous polymer as the skin-forming component can suppress bubble growth, the final product is expected to have better uniformity in cell size and its distribution and hence improved mechanical properties (e.g., tensile and flexural moduli).

On the basis of the photomicrographs and the tensile properties taken of extrudate samples, it can be concluded that the quality of foam (i.e., the cell size and its distribution in the foamed core) may be controlled by judiciously adjusting the thickness ratio of the skin to core components and the viscosity ratio of the skin to core components.

This work is partly taken from the dissertation of Ramakrishna Shetty, submitted to the Faculty of the Polytechnic Institute of New York in partial fulfillment of the requirements for the degree of Doctor of Philosophy, 1977.

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