

Basic Study of Extrusion of Polyethylene and Polystyrene Foams

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Synopsis

A fundamental study of bubble morphology development and apparent rheological properties in foam extrusion is reported. The influence of melt temperature, die length/diameter ratio, and blowing agent level on the morphology are considered. Measurements of the influence of blowing agent on viscosity, extrudate swell, and end-pressure losses are described. The viscosity is reduced, but extrudate swell is increased. End-pressure losses were found to become very large relative to the die wall shear stress at low extrusion rates. These results were interpreted in terms of bubble development. The filling of molds by foaming melts was observed and is described.

INTRODUCTION

Extrusion and injection molding of foams are important industrial plastics processing operations.¹⁻⁷ Future improvements in these processes may well depend on a better fundamental understanding of the process and mechanisms involved. Relatively few basic studies of the formation of thermoplastic foams have appeared in the literature. We present such a study here.

Studies of foam extrusion have been reported by Hansen¹ Blyler and Kwei,⁸ Bigg et al.,² and Han et al.⁹ The only basic investigations of injection molding of foams are apparently those published by Throne^{6,7} and Gross and Angell.¹⁰ An important aspect of the formation of foams is the mechanics of bubble growth in polymer melts. There are few experimental studies of growth in the literature, and one can only note those of Denecour and Gent¹¹ and Gent and Tompkins,¹² who have investigated bubble growth in elastomers. We are aware of experimental studies currently being carried out by Han and his coworkers¹³ on polymer melts. There have been numerous theoretical studies of bubble formation generally in inviscid or Newtonian fluids.¹⁴⁻¹⁶ Yang and Yeh¹⁷ have investigated bubble growth in viscous non-Newtonian fluids, whereas Street and his coworkers^{18,19} have presented analyses for viscoelastic fluids. Street et al.¹⁹ and Hobbs²⁰ have attempted to use these theories of bubble growth to interpret the formation of polymeric foams.

In this paper, we present an experimental study of the flow of foaming polymer melts through a die. Some observations of mold filling are reported. We investigate variations viscosity, end-pressure losses and extrudate swell. Extrudate cross sections are investigated using scanning electron microscopy. We look at the influence of temperature, residence time, and melt viscosity on bubble development. The influence of these variables on apparent viscosity and end-pressure losses is studied. The mode of mold filling and morphology of molded parts will be investigated.

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EXPERIMENTAL

Materials. Various polyethylenes and polystyrenes were used in this study and are summarized in Table I. They were chosen so as to represent a range of melt rheological properties. Celogen AZ-130 (azodicarbonamide) blowing agent supplied by Uniroyal was used in this study.

Mixing. The blowing agents were mixed into the polymers in a 0.75-in. bender screw extruder operating at 140°C. The blowing agents were mixed into each of the polymers in varying ratios. In the case of LDPE-2 and PS-2, this was in the proportions 0, 0.5, 1.0, and 1.5%.

Extrusion. The plastic/foaming agent blends were extruded at different temperatures (170, 190, 210°C) from an Instron Capillary Rheometer. The blends were heated for a period of 10 min before extrusion. Dies of diameter 1.46 mm and length/diameter (L/D) ratios 5, 25, and 40 were used. The total pressure drop, P_T , between the reservoir and the atmosphere was resolved between a die-pressure loss and an ends-pressure loss.

The pressure loss may be related to the wall shear stress $(\sigma_{12})_w$, i.e.,²³⁻²⁶

$$P_T = \Delta P_{\text{die}} + \Delta P_{\text{end}} \quad (1)$$

$$= 4(\sigma_{12})_w \frac{L}{D} + \Delta P_{\text{end}} \quad (2)$$

where ΔP_{end} should be considered as the sum of entrance- and exit-pressure losses.

The shear rate at the die wall for a homogeneous melt is given by Weissenberg's relation²⁴

$$\dot{\gamma}_w = \left(\frac{3n' + 1}{4n'} \right) \frac{32Q}{\pi D^3} \quad (3)$$

where

$$n' = d \log(\sigma_{12})_w / d \log 32Q/\pi D^3$$

Injection Molding. Injection molding with flow visualization (Fig. 1) was carried out using a mold with glass windows attached to the Instron Capillary Rheometer. This is essentially the apparatus described by White and Dee²⁷ and by Oda et al.²⁸ The melt temperatures were 190°C and the mold temperature was varied from 110 to 170°C.

Void Fraction. Void fractions of extrudates were obtained by measuring the relative masses of foamed and solid extrudates of known dimensions.

TABLE I
Polymers Investigated in Structural Foam

Polymer	Source	Designation	Molecular and rheological characterization
Polystyrene	ARCO experimental polymer	PS-1	High viscosity, experimental sample
	Dow styron 6780	PS-2 ^a	White and Roman ¹⁹
Low-density polyethylene	Tenite 1811F	LDPE-1	MI = 0.7
	Dow tyon 610M	LDPE-2 ^a	MI = 5.0, White and Roman, ²¹ Ide and White ²²
	DNDA 0455	LDPE-3	MI = 60
High-density polyethylene	Marlex EMB6050	HDPE	MI = 5.0, White and Roman ²¹

^a Main materials.

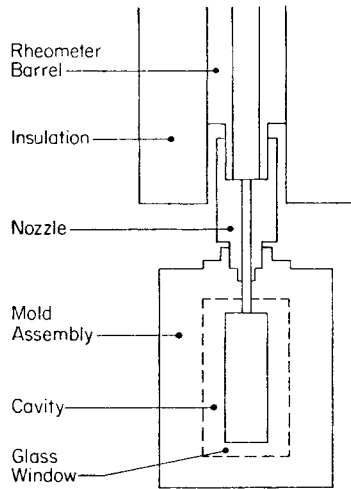


Fig. 1. Injection molding flow visualization apparatus.

Scanning Electron Microscopy. SEM photomicrographs were made on cross sections of foamed extrudates and foamed injection-molded parts. The studies were carried out using an AMR Model 900 high-resolution scanning electron microscope (Advanced Metals Research Corporation, Burlington, Mass.). A gold-palladium alloy was used on the specimens to avoid charging in the electron beam. Generally, the cross section of parts to be placed in the SEM were prepared by quenching in liquid nitrogen bath and fracturing. Mean diameters of bubbles \bar{d} and frequency of occurrence of bubbles \bar{n} per unit area were determined from the SEM photomicrographs.

RESULTS

Extrudate Cross Sections

SEM photomicrographs of cross sections of extrudates were obtained. Representative cross sections for the LDPE-2 with 1% blowing agent at 170, 190, and 210°C are shown in Fig. 2. These were extruded from a 40 L/D die. Generally, extrudates foamed at 170°C are relatively solid, except for HDPE. The extrudates foamed at 190 and 210°C possess bubbles throughout their cross section. Thus, there is at first a tendency for bubble frequency to increase with temperature. However, bubble sizes become larger with rising temperature and this then leads to a decrease in the frequency.

The effect of blowing agent level is shown in Fig. 3. The influence of die L/D ratio on extrudate cross section for the melt at 190°C is given in Fig. 4.

It is seen that the larger the L/D ratio, the smaller the bubble size under a constant extrusion rate condition.

Characterization of Extrudate Porosity

We have measured void fractions, ϵ , and bubble diameters in the extrudates. Significant variations are found with changes in temperature and extrusion rate for any particular polymer. There seems to be only minor or complex effects

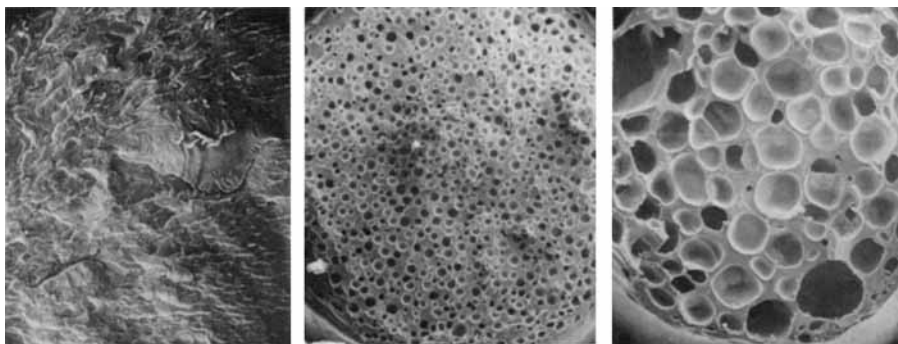


Fig. 2. SEM photomicrographs of the LDPE-2 extrudate cross sections extruded at 170 (left), 190 (center), and 210°C (right). $L/D = 40$ and 1% AZ in all cases. Left, $\times 100$; center and right, $\times 50$.



Fig. 3. SEM photomicrographs showing the influence of 0.5 (left), 1.0 (center), and 1.5% blowing agent (right) on the extrudate cross section of PS-2 extrudate foam at 190°C. $L/D = 25$; shear rate $\dot{\gamma}_w = 50 \text{ sec}^{-1}$. The magnification is $\times 50$ in all cases.

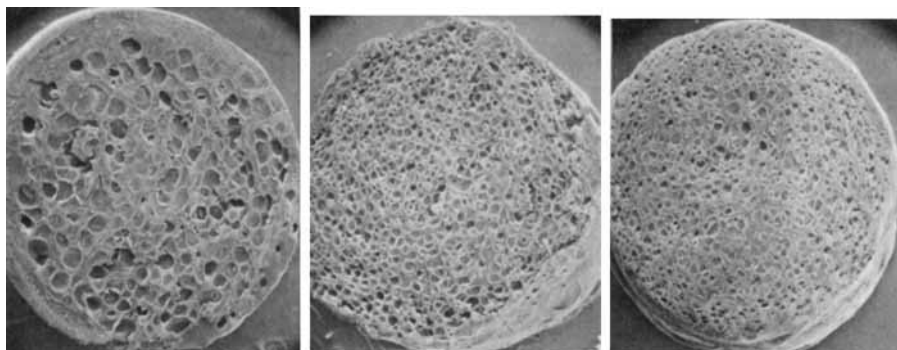


Fig. 4. SEM photomicrographs of the PS-2 extrudate cross sections foamed at 190°C, where $L/D = 5$ (left, $\times 42$), 25 (center, $\times 40$), and 40 (right, $\times 50$). $\dot{\gamma}_w = 50 \text{ sec}^{-1}$ and 0.5% AZ in all cases.

with blowing agent level. Figures 5 and 6 show occupied fraction ($1 - \epsilon$) and mean bubble diameter, \bar{d} , as a function of die wall shear rate and residence time for PS-2 at 170, 190, and 210°C and LDPE-1 at 190°C. It is clear that the void fraction increases with temperature and decreasing extrusion rate. The bubble diameters similarly increase with both temperature and decreasing extrusion rates. Large residence times are equivalent to low shear rates in any particular

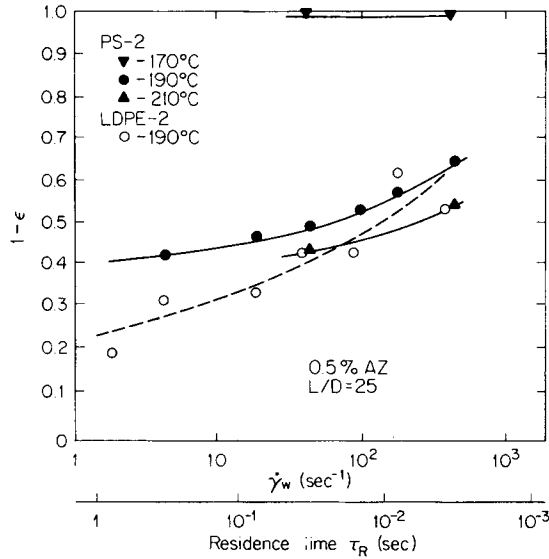


Fig. 5. Occupied fraction of $1 - \epsilon$ of foamed extrudate as a function of die wall shear rate and residence time for the PS-2 at 170, 190, 210°C and the LDPE-2 at 190°C.

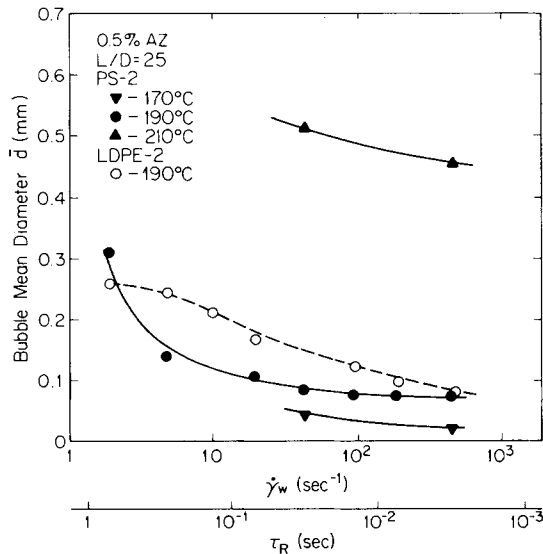


Fig. 6. Mean bubble diameter per unit area as a function of die wall shear rate and residence time for the PS-2 at 170, 190, and 210°C and the LDPE-2 at 190°C.

die. Void fractions and bubble diameters thus seem to increase with die residence time.

Extrudate Swell

In Figs. 7 and 8 we plot extrudate swell, B , equal to (extrudate diameter)/(die diameter), as a function of shear rate for the melts studied at $L/D = 25$. The plots include data with and without blowing agent.

Generally, extrudate swell in the absence of blowing agent increases with shear rate and with decreasing temperature.

The presence of blowing agent increases extrudate swell at 190 and 210°C.

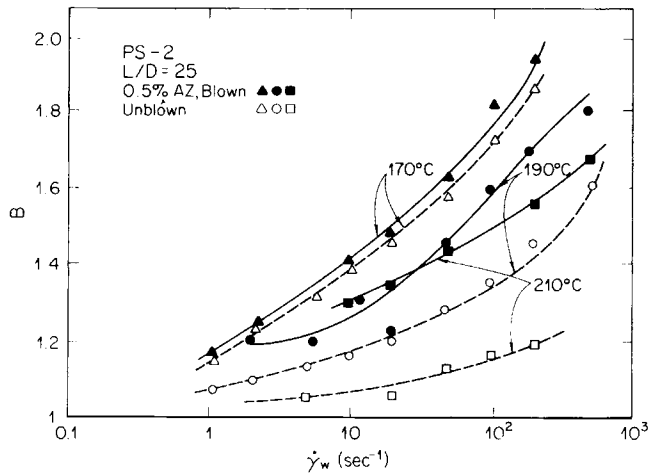


Fig. 7. Extrudate swell of the PS-2 at $L/D = 25$ as a function of wall shear rate at 170, 190, and 210°C, with and without blowing agent.

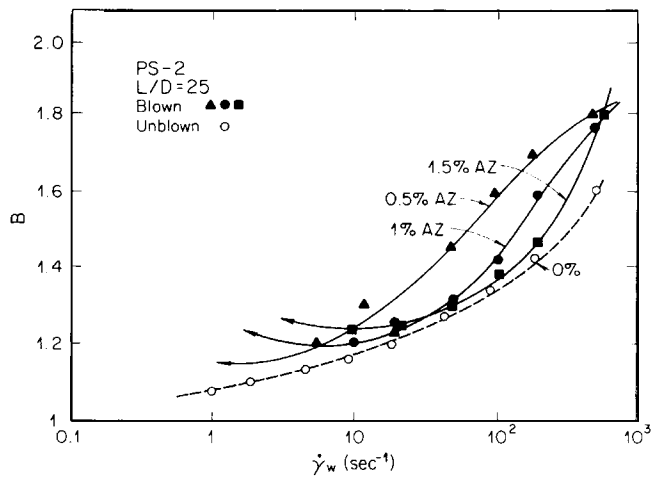


Fig. 8. Extrudate swell of the PS-2 at $L/D = 25$ as a function of wall shear rate at 190°C with blowing agent levels of 0, 0.5, 1.0, and 1.5%.

Figure 7 shows the influence of temperature and Fig. 8 the effect of the level of blowing agent. The behavior is obviously complex. Strikingly, addition of 0.5% blowing agent has a greater effect in increasing swell than the 1.0 and 1.5% levels.

Total Extrusion Pressure

The total pressure P_T is plotted as a function of the level of blowing agent in Fig. 9. It is seen that P_T rises steeply at low shear rates, but mildly decreases at higher shear rates. Generally, P_T at low shear rate shows a constant value at 190 and 210°C.

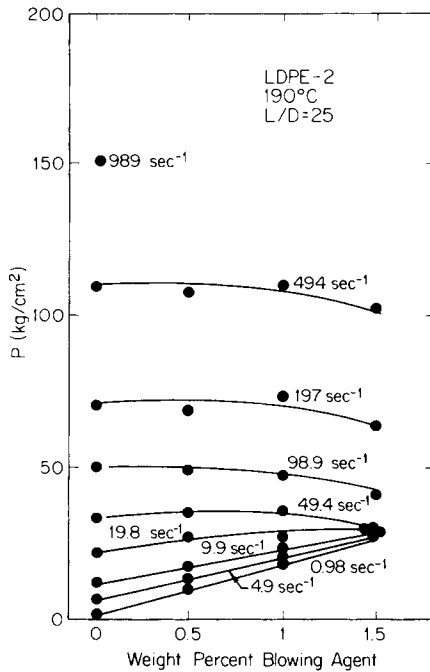


Fig. 9. Total pressure P_T as a function of blowing agent (wt %) for the LDPE-2 at 190°C for various $\dot{\gamma}_w$ in $L/D = 25$ die.

Viscosity Behavior

In Figs. 10–12 we plot the *apparent* viscosity of three polymer melts at 170, 190, and 210°C. The figures contain data for the melts with and without blowing agent. Generally, the viscosity decreases with increase in temperature and with increasing shear rate. At 170°C the viscosity is the same for the melt with and without the blowing agent. At 190 and 210°C, the viscosity of the system with blowing agent is lower.

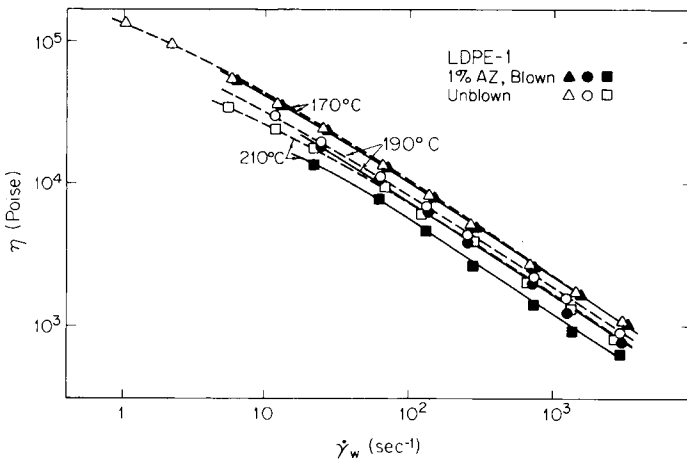


Fig. 10. Apparent viscosity as a function of shear rate for the LDPE-1 at 170, 190, and 210°C, with and without blowing agent.

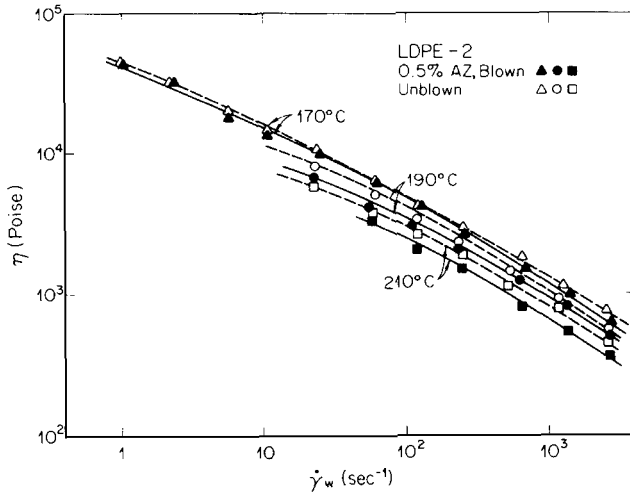


Fig. 11. Apparent viscosity as a function of shear rate for the LDPE-2 at 170, 190, and 210°C, with and without blowing agent.

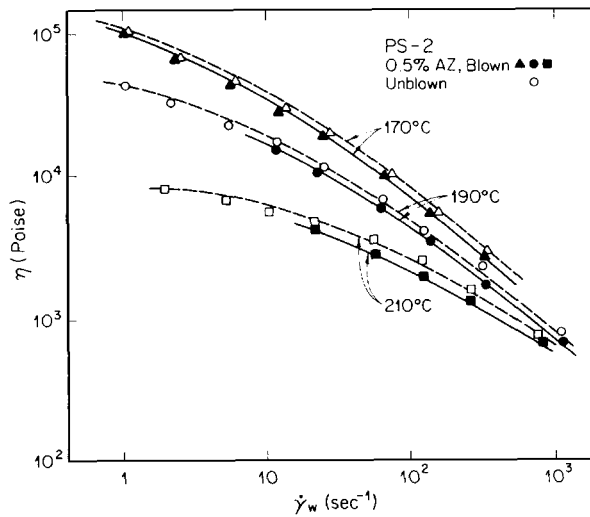


Fig. 12. Apparent viscosity as a function of shear rate for the PS-2 at 170, 190, and 210°C, with and without blowing agent.

End-Pressure Losses

End pressure losses normalized with die wall shear stress are plotted as a function of capillary wall shear rate in Figs. 13 and 14. At 170°C the data for systems with and without blowing agents are the same. This is not the case at 190 and 210°C. In Fig. 13 the large values of $\Delta P_{\text{end}}/(\sigma_{12})_w$ at low shear rates for the LDPE-2 with 0.5 and 1.07% blowing agent are apparent. In Figure 14 we see that the PS-2 with blowing agent generally has larger values of $\Delta P_{\text{end}}/(\sigma_{12})_w$.

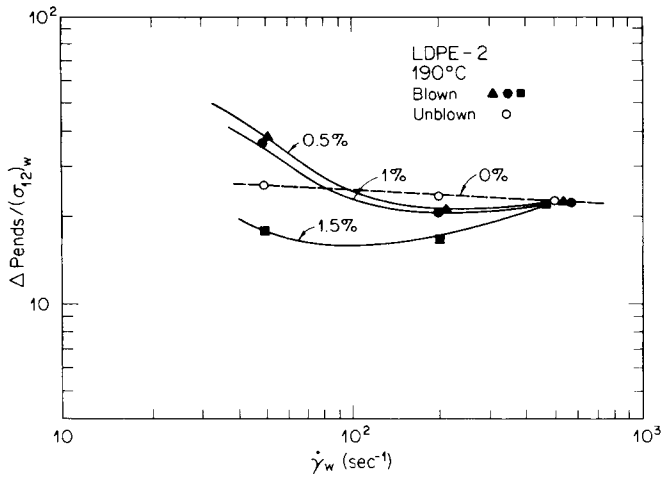


Fig. 13. End-pressure loss reduced with shear stress $\Delta P_{end}/(\sigma_{12})_w$ as a function of shear rate at 190°C with different levels of blowing agent.

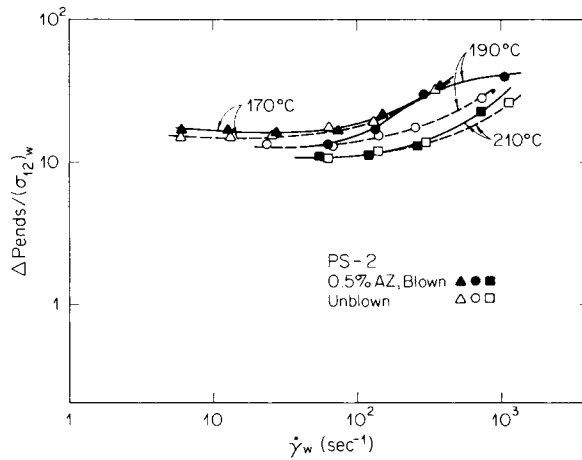
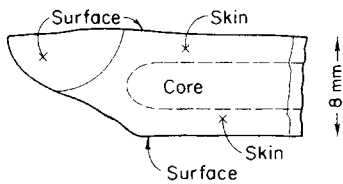


Fig. 14. End-pressure loss reduced with shear stress $\Delta P_{end}/(\sigma_{12})_w$ as a function of shear rate for the PS-2 at 170, 190, and 210°C, with and without blowing agent.

Injection Molding

Flow visualization of the injection mold filling was carried out under nonisothermal conditions. The melt was observed to be opaque as it exited the runner and entered the mold. Generally, mold filling occurred by normal front development as described in our earlier papers.^{27,28} The melt adheres to the glass windows of the mold and flows through the mold in patterns resembling Hele-Shaw flow.²⁹ If a large-thickness mold insert is used, i.e., mold channel depth is increased relative to gate diameter, the melt is observed to initially jet into the mold. However, the jet continues to expand within the mold walls and eventually contacts them. The jetting regime evolves into the simple or normal mold-filling behavior.

The cross sections of injection molded parts were examined using a scanning electron microscope. An SEM stereo scan of a cross section of a molded part is shown in Fig. 15. The bubbles may be seen in the cross section. The effects



Moldings Dimension: 74 x 25 x 8 mm

Molding Condition;

Melt Temp. 190°C

Mold Temp. 130°C

Injection Speed 20 in/min

Density (Average) 0.69

Blow Ratio 1.35

Fig. 15. SEM photomicrograph stereos of injection molded foam part for the LDPE-2.

of injection speed and mold temperature on the surface roughness is relatively large. High-viscosity polymers with blowing agent are more difficult to process because of the high processing pressure.

DISCUSSION

Extrusion

The experimental results on extrusion are generally compatible with the earlier studies of Blyler and Kwei⁸ and Han et al.⁹ Apparent viscosities are reduced by the presence of foaming agents, and presumably similar associated bubble development is found. One of the reasons for the viscosity decrease of the melt with blowing agent is the decreasing of melt volume in a die by bubbles. The influence of the gas bubbles on the velocity field is not known. It might reasonably be surmised that the bubbles are primarily at large radii near the die wall, where the residence time is higher. This might help explain the lowered viscosity.

Complex pressure loss effects in extrusion are found. P_T increases with blowing agent level at low extrusion rates despite a reduction in viscosity. $\Delta P_{\text{end}}/(\sigma_{12})_w$ tends to increase with blowing agent, especially at low extrusion rates. The implications of this may be expressed in an interesting manner by considering a viscosity function, η^* , defined in terms of P_T rather than ΔP_{die} , i.e.,

$$\eta^* = (DP_T/4L)/\dot{\gamma}_w \quad (4)$$

This is shown in Figs. 16–18. At low $\dot{\gamma}_w$, η^* becomes very large and continues to increase with decreasing $\dot{\gamma}_w$. In Fig. 19, we plot the critical point of upturn of the viscosity curve for LDPE-2 as a function of blowing agent, die L/D , and

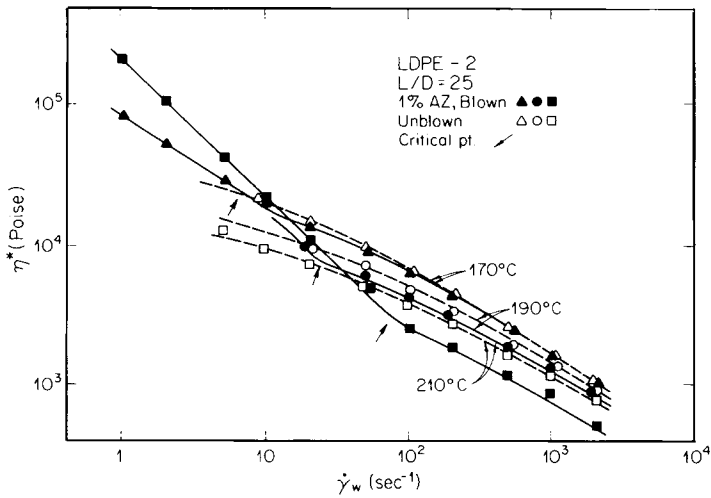


Fig. 16. Apparent viscosity, η^* , of eq. (4) based on P_T as a function of $\dot{\gamma}_w$ for the LDPE-2 containing 1.0% blowing agent at 170, 190, and 210°C.

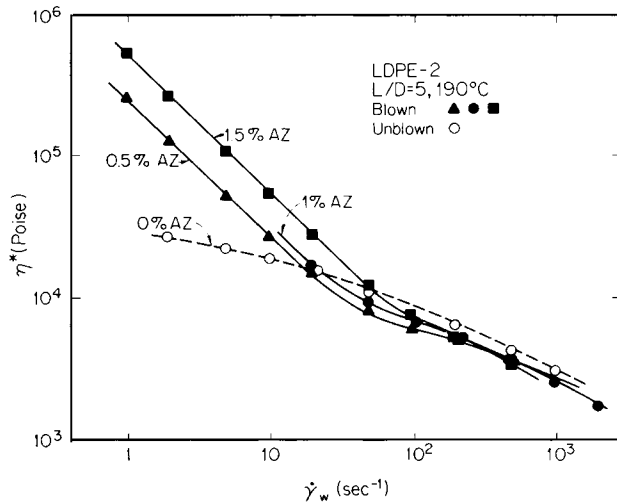


Fig. 17. η^* as a function of $\dot{\gamma}_w$ for the LDPE-2 at 190°C with different levels of blowing agent.

temperature. The critical shear rate increases with blowing agent content and temperature and decreases with increasing die L/D ratio.

It is of interest to compare our results with those of the excellent study of Han et al.,⁹ who have measured pressure profiles along the length of a die in foam extrusion. The end-pressure loss is a sum of an entrance and an exit loss:

$$\Delta P_{\text{end}} = \Delta P_{\text{ent}} + \Delta P_{\text{exit}} \tag{5}$$

Han et al. find that the pressure of the foaming agent increases ΔP_{ent} , but decreases ΔP_{exit} substantially. This clearly means that the very large values of η^* at low $\dot{\gamma}_w$ are due to ΔP_{ent} , and indeed using ΔP_{end} underestimates the effect.

There is a strong correlation of the high values of η^* and the critical shear rates with the same parameters that determine bubble size. Large bubbles are found

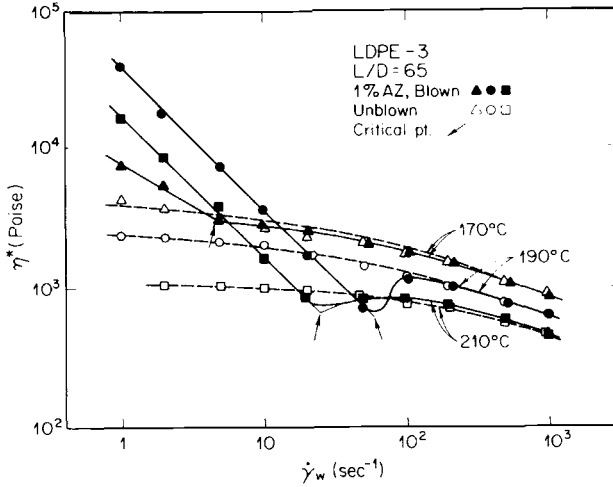


Fig. 18. η^* as a function of $\dot{\gamma}_w$ for the LDPE-3 at 170, 190, and 210°C, with and without blowing agent.

with low $\dot{\gamma}_w$ and high temperatures. The large ΔP_{end} and gas bubble size may be associated with greater tendencies for bubbles to form in the reservoir rather than the die when the pressure is low. Presumably, the large bubbles considerably disrupt flow behavior in the converging flow at the die entrance. This gives rise to excessive entrance-pressure losses.

Injection Molding

The general observations of mold filling are in agreement with the earlier work of Oda et al.²⁸ if we accept the idea that in a hot mold a foamed extrudate will continue to increase in diameter due to bubble growth in the melt. Jetting to the end of the mold is possible only if the time required for the melt to reach the end of the mold is less than that required for the swelling extrudate to contact the mold walls. The above idea can be expressed in analytical terms as

$$t_{mold, res} = \int_0^{L_{mold}} \frac{dx}{v_j(x)} > \int_{B(0)}^{h/D} \frac{dB(t)}{dB/dt} \tag{6}$$

where x is the coordinate reaching from the gate to the end of the mold, $B(t_{mold, res})$ is the die swell as a function of residence time in the mold, v_j is the velocity of the jet, and h is the narrow cross section of the mold. We need to note

$$v_j(x) = \frac{4Q}{\pi d^2} = \frac{4Q}{\pi D^2} \frac{1}{B^2} \tag{7}$$

where Q is the extrusion rate and d is the extrudate diameter. To evaluate eq. (6), we need to know $B(t)$.

CONCLUSIONS

The results obtained by the bubble morphology and rheological behavior in foam extrusion may be summarized as follows. Bubble size \bar{d} and void fraction

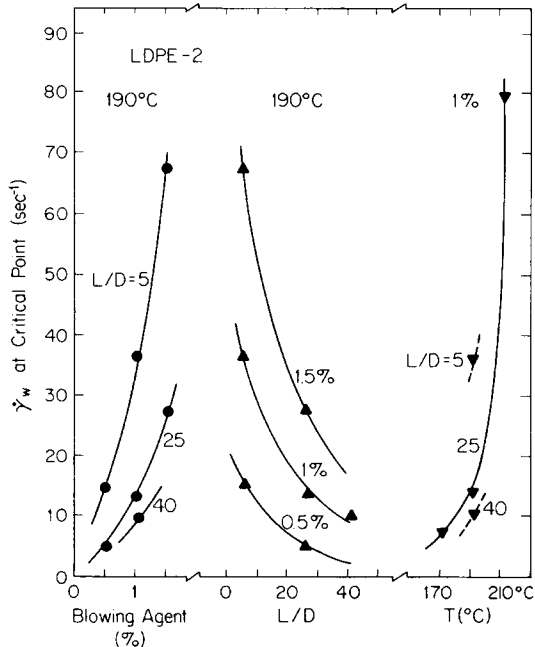


Fig. 19. The critical point of upturn of the viscosity curve for the LDPE-2 as a function of blowing agent levels, die L/D , and temperature.

ϵ increases with (1) temperature, (2) decreasing extrusion rates, and (3) decreasing die L/D ratio (under constant extrusion rate conditions).

Bubble size generally correlates inversely with melt pressure.

The extrudate swell B of melts containing blowing agent is greater than melts without such additives. For the foamed extrudates, B increases with the shear rate and decreasing temperature, but the effect of blowing agent level on the swell is obviously complex. The viscosity of the melts with blowing agent is smaller than that without blowing agent, especially at high temperatures. The effect of blowing agent on the total pressure required for extrusion is greatest at low shear rates.

The anomalous extrusion-pressure effects found are associated with the presence of large bubbles, and probably with bubble formation in the reservoir.

Studies of mold filling by foams are described. Special attention is given to the problem of jetting.

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References

1. R. H. Hansen, *SPE J.*, **18**, 77 (1962).
2. D. M. Bigg, J. B. Preston, and D. Brenner, *Polym. Eng. Sci.*, **16**, 706 (1976).
3. H. Gonzalez, *J. Cell. Plast.*, **12**, 49 (1976).
4. B. S. Mehta and E. A. Columbo, *J. Cell. Plast.*, **12**, 98 (1976).
5. S. Y. Hobbs, *J. Cell. Plast.*, **12**, 258 (1976).

6. J. L. Throne, *J. Cell. Plast.*, **12**, 161 (1976).
7. J. L. Throne, *J. Cell. Plast.*, **12**, 264 (1976).
8. L. L. Blyler and I. W. Kwei, *J. Polym. Sci., Part C*, **35**, 165 (1971).
9. C. D. Han, Y. W. Kim, and K. D. Malhotra, *J. Appl. Polym. Sci.*, **20**, 1583 (1976).
10. L. B. Gross and R. G. Angell, *Soc. Plast. Eng. ANTEC. Tech. Pap.*, **34**, 162 (1976).
11. R. L. Denecour and A. N. Gent, *J. Polym. Sci., Part A*, **6**, 1852 (1968).
12. A. N. Gent and P. A. Tompkins, *J. Appl. Phys.*, **40**, 2520 (1969).
13. C. D. Han, unpublished research, 1977.
14. H. Lamb, *Hydrodynamics*, 6th ed., Dover, New York, 1944.
15. M. Plesset, *J. Appl. Mech.*, **16**, 277 (1949).
- 15a. M. Plesset and S. A. Zwick, *J. Appl. Phys.*, **25**, 499 (1954).
16. E. L. Barlow and W. E. Langlois, *IBM J. Res. Dev.*, **6**, 329 (1962).
17. W. J. Yang and H. C. Yeh, *AIChE J.*, **12**, 927 (1966).
18. J. R. Street, *Trans. Soc. Rheol.*, **12**, 103 (1968).
19. J. R. Street, A. L. Fricke, and P. Reiss, *IEC Fund. Res. Dev.*, **10**, 54 (1971).
20. S. Y. Hobbs, *Polym. Eng. Sci.*, **16**, 270 (1976).
21. J. L. White and J. F. Roman, *J. Appl. Polym. Sci.*, **20**, 1005 (1976).
22. Y. Ide and J. L. White, *J. Appl. Polym. Sci.*, **22**, 1061 (1978).
23. E. B. Bagley, *J. Appl. Phys.*, **28**, 624 (1957).
24. T. F. Ballenger and J. L. White, *J. Appl. Polym. Sci.*, **15**, 1949 (1971).
25. J. L. White, in U.S.—Japan Seminar, Polymer Processing and Rheology, edited by D. C. Bogue,
- M. Yamamoto and J. L. White, *Appl. Polym. Symp.*, **20**, 155 (1973).
26. S. Middleman, *The Flow of High Polymers*, Wiley, New York, 1968.
27. J. L. White and H. B. Dee, *Polym. Eng. Sci.*, **14**, 212 (1974).
28. K. Oda, J. L. White, and E. S. Clark, *Polym. Eng. Sci.*, **16**, 585 (1976).
29. J. L. White, *Polym. Eng. Sci.*, **15**, 44 (1975).

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