SOCIETY OF PLASTICS ENGINEERS REGIONAL TECHNICAL CONFERENCE October 11-13, 1993

Injection Molding Outlook

Co-Sponsors: North Texas Section and Injection Molding Division

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LAMELLAR INJECTION MOLDING

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INTRODUCTION

Lamellar Injection Molding (LIM) is patented fabrication technology which can produce net shape parts having a lamellar blend morphology (1 - 4). LIM combines coextrusion and injection molding technology to enable the molder to create a micron scale layered blend morphology in complex asymmetric multicavity parts (2). Lamellar blends based on two or more polymers can enhance such properties as gas and solvent barrier (3), chemical and temperature resistance (4), coefficient of thermal expansion, and optical clarity (4), relative to a conventional blend analogs which assume a globular or columnar phase morphology due primarily to thermodynamic and rheological considerations (5).

Coextrusion is a well established technology for the fabrication of various structures (e.g., barrier packaging), and it is ideally suited for simple articles that can be extrusion blowmolded or thermoformed. The traditional three layer sandwich coinjection molding process is not amenable to molding multicavity parts, or complex asymmetric shapes, as compositional uniformity cannot be maintained (both intracavity and intercavity). Recent developments are claimed to enable multicavity coinjection molding, however, these suffer from either (a) extremely complex process control and coinjection nozzle design (6,7), or (b) expensive mechanically complex molds that separately meter individual components into each individual mold cavity (8).

LIM circumvents the inherent limitations of traditional coinjection molding by using simultaneous injection coupled with layer multiplication to produce a finely subdivided (micron scale) multilayer meltstream incident to the mold. LIM creates a layered melt stream within the molding machine, and lamellar morphology can be retained in complex multicavity parts even with a minor component as low as 2% (2). LIM can be used with conventional molds and standard injection molding process controls. Existing multibarrel injection presses can be retrofitted for LIM.

The property enhancements resulting from the lamellar morphology provided through LIM can equal those of multilayer coextrusion and coinjection molding (3). Gas permeability, which has a well understood structure-property relationship (9), can be used as an example to illustrate the value of LIM technology. Theoretical predictions of oxygen transmission rate for two morphological extremes (random discontinuous morphology and perfect lamellar morphology) are compared in *figure 1* for linerar polyethylene - EVOH blends. The data for LIM approaches the ideal layered structure normally realized in coextrusion, and with only 8% EVOH a 300 fold reduction in permeability is realized relative to conventional blends.

This paper describes the LIM process as practiced in developmental scale-up using a 715 ton dual barrel reciprocating screw injection molding machine.

LIM PROCESS

Initia! laboratory work to demonstrate LIM used continuous coextrusion coupled with a modified transfer molding process that simulated the mold fill step of injection molding, i.e., fountain flow was realized at the free flow front during mold fill where high extensional strain rates can occur (10). The transfer molding process successfully demonstrated the viability of maintaining substantially continuous lamella with good compositional uniformity (2). Three component LIM was demonstrated, and it was shown that adhesive (or compatibilizing) layers could be retained at the interfaces between incompatible polymer pairs (3).

Since this initial work used multilayer coextrusion to produce a multilayer melt stream, new technical challenges were presented when the process was applied to reciprociating screw injection molding.

- 1. Intermittent flow and high instantaneous flow rates (e.g., 1000 kg/hr) and pressures (1300 bar).
- 2. Intermittent layer generation at shear rates and stresses much higher (ca. 100x) than those experienced during continuous coextrusion.

 Compositional control determined by relative injection rates rather than precision metering by gear pumps or gravimetric feeders.

A schematic of the LIM process as practiced in developmental scale-up is shown in *figure 2*. Two injection cylinders are used to generate individual meltstreams of components A and B. Injection of the two components occurs simultaneously at predetermined rates governed by the desired composition ratios. The individual melt streams are then combined in a feedblock to arrange a three layer structure (A-B-A). The combined stream then passes through a series of layer multipliers which repeatedly subdivide and stack the layers to increase number and reduce thickness. The process can be repeated several times, and the resultant total number of layers for a symmetrical structure is given by *equation 1*:

$$M = 4^{N}(n-1) + 1 \tag{1}$$

where: M = Total number of layers.

n =Initial number of layers.

N = Number of layer multiplication stages.

The melt stream typically passes through a standard 1cm diameter injection nozzle prior to mold filling. While developmental scale-up was done using a two injection cylinder machine, the process is believed suitable for using three injection units and a five layer feedblock. This allows use of an adhesive polymer to bond non-adhering polymers. The controlled placement of an adhesive or compatibilizing polymer at the interfaces represents a further advantage of LIM over conventional blend technology. Additional injection units could be incorporated if desired.

The machine chosen for the developmental scale-up phase for LIM was a 6500 kN (715 U.S. tons) Battenfeld dual barrel injection molding machine which is illustrated pictorially in *figure 3*. The original coinjection hardware was replaced with a three layer feedblock and layer generation stage as shown in the *figure 2* schematic and pictorially in *figures 3* and *4*. Each layer multiplier subdivides the incident meltstream into four substreams, which are subsequently reoriented, stacked, and recombined (11). A modular design was used to permit relative ease of changing number of layers. The number of feedstream layers can be varied in increments of 3, 9, 33, 129, 513, and 2049 in the practiced configuration. Other layer combinations can be obtained by using either two or three channel layer multipliers, or a feedblock with more than three layers. Retrofit was accomplished without significant modification to either the control system or machine.

Figure 5 illustrates typical nozzle morphologies observed using the Battenfeld injection press equipped with three layer multiplication stages to give 129 feedstream layers. The materials used were pigmented high impact polystyrene (HIPS) resins. Figure 5a is a steady state extrudate processed at an apparent shear rate of 980 sec⁻¹. Figure 5b is an injected air shot processed at an instantaneous apparent shear rate of 4500 sec⁻¹. Both materials exhibit lamellar morphology. Differences observed between extrusion and injection are attributed to (1) higher shear rates, and (2) the intermittant nature of the layer multiplication process when practiced using simultaneous injection.

The three primary molds evaluated to date are described below:

- 1. A 16 in long bumper beam prototype mold. It can be operated as a single cavity or two cavity mold, with either single or multiple (up to 3) gates per cavity.
- 2. A 13" TV cabinet mold that is single gated. The flow path is extremely complex and contains multiple weld lines, particularly at the vent regions. Work with this mold confirmed that substantial lamella retention could be maintained throughout a tortuous flow path that contained multiple weld lines (2).
- 3. A 16 cavity mold that simulates multicavity molding using common balanced and unbalanced runner configurations (shown schematically in *figure 6*).

The LIM process has been found to be extremely repeatable when practiced by following generally accepted engineering principles (2). An example of this is given in *figure 7*, which illustrates a series of cross-sectional optical photomicrographs of parts obtained during a 50 cycle run of pigmented HIPS that employed the bumper beam mold. Eight randomly selected samples were evaluated for internal

morphology. Near fingerprint quality replication was observed which exceeded expectations. Process data for this study is given in *figure 8*.

Numerous two and three component LIM systems have been evaluated using the laboratory transfer molding process, and several of these have been studied for proving developmental scale-up using the dual barrel injection molding process (2). Part performance data are given elsewhere (3,4).

PROCESS AND DESIGN CONSIDERATIONS

Two conditions must be satisfied for the successful practice of LIM. Specifically, the molded article must maintain (a) substantial compositional uniformity, and (b) substantial layer continuity throughout the part.

Compositional uniformity hinges on maintaining constant flow rates during simultaneous injection of the components, maintaining axial and lateral layer uniformity throughout the layer generation process, and having a minimum number of feedstream layers to ensure adequate compositional uniformity throughout the molded article. The former involves control of the injection cycle to maintain a constant volumetric ratio of components to the feedblock. Layer uniformity involves proper design of the feedblock and layer multipliers so that all portions of the lamellar melt stream have nearly the same composition. This assures that irrespective of which portion of the melt stream fills the mold cavities, good uniformity of composition is maintained if an adequate number of feedstream are initially present.

Layer continuity is defined by a critical lamella thickness which can initiate layer breakup. The average layer thickness for a given volumetric composition decreases with increasing number of layers. A large number of relatively thin layers is desired to ensure compositional uniformity in all portions of the mold. A simple simultaneous injection of a three layer melt stream will display compositional non-uniformity because of solidification of the outer layers on the mold wall during mold fill. A microlayer melt stream will fill the mold with a relatively constant composition as lamella continue to fill the mold during fountain flow with simultaneous solidification. The minimum layer thickness is defined by relative rheological properties, interfacial tension, and flow kinematics. A reasonable viscosity match of polymer components would be to within a factor of about three. Low interfacial tension between polymers is preferred, and streamlined symmetrical flow paths and minimum stresses during layer generation are desired.

Figure 9, gas permeability versus number of feedstream layers, illustrates the general permeability process-structure-property relationship observed in LIM articles containing small quantities of barrier polymer. Four fundamental zones are evident, which relate to the aforementioned conditions:

Zone 1 (decreasing permeability): An insufficient number of layers results in a high permeability due to non-uniform material distribution.

Zone 2 (lower bound permeability): Material distribution is uniform, lamella are continuous, and the theoretical prediction (9) for a lamellar structure is realized over an operating window that is material, composition, and process dependent.

Zone 3 (increasing permeability): Lamella begin to reach a critical thickness that initiates spontaneous breakup and coalescence into discontinuous domains.

Zone 4 (upper bound permeability): The initial lamellar morphology has been completely erased, and the theoretical prediction (9) for a random dispersion is followed.

Figure 10 Illustrates these regions for LIM structures based on HDPE - Adhesive - EVOH (8 vol% EVOH). A constant lower bound permeability zone, which represents a 300X improvement relative to a simple blend, exists over a broad operating window; 65 to 1025 feedstream layers. Lamella breakup begins to occur at average barrier layer thickness ranging from 0.1 μ m - 0.3 μ m, and the upper bound is approached as lamella thicknesses become diminishingly small.

The breadth of the LIM operating window, defined as points a and b in figure 9, will be defined by the following factors:

Minimum Number of Lavers (a)

- Composition Ratio
- Number of Cavities
- Flow Path Length
- Mold Complexity

Maximum Number of Lavers (b)

- Composition Ratio
- Rheological Properties
- Interfacial Tension
- Flow Kinematics
- Tool and Gate Design

Work to date, using various LIM systems (ca. 10% barrier polymer) with a single cavity 1 mm thick can mold, has established approximately 10 to 20 feedstream layers as the minimum. Multicavity molding will shift this number upward; the specific magnitude will be defined by the number of cavities, runner configuration, and tool design.

Gate and Runner Design: The LIM process has been evaluated with several gate designs, specifically:

(a) tab, (b) submarine, and (c) modified fan. No differences have been seen between the different gate configurations at shear rates approaching 30,000 sec⁻¹. Pin gates have not been evaluated to date, however, layer retention would be anticipated if appropriate engineering considerations are employed. Hot runners have also not been evaluated, but first principles suggest that LIM would be amenable to hot runner molding, and most likely the preferred runner system for practicing the technology, as scrap rates would be reduced.

<u>Wall Thickness Effects</u>: Structures evaluated by LIM to date have ranged in thickness from approximately 1 mm to 6 mm thick. Thinner structures could be produced by following prudent engineering principles. Average layer thickness, the critical parameter which determines the onset of layer breakup, is largely material dependent. For a given material combination and composition, part thickness will dictate the maximum number of feedstream layers permissible for LIM.

Design equations can be derived to estimate the maximum permissible number of layers, given the material composition, part thickness, and layer breakup parameter (l_c) which is defined as the average lamella thickness of the minor component that initiates breakup and subsequent coalescence. These design equations are given below for two $(equation\ 2)$ and three $(equation\ 3)$ component structures, where the minor component is assumed as (a) present as the center (core) layer in the feedblock, and (b) the component which initiates the layer breakup process. This analysis assumes that kinematics of mold fill is a constant, and therefore serves only as a guide for estimate purposes only.

$$M_p \approx 2 \left(\phi h / l_c \right)$$
 (2)

$$M_{\rm D} \approx 4 \left(\phi h / l_{\rm C} \right) \tag{3}$$

where: M_D = Maximum permissible number of feedstream layers.

 \emptyset = Volume fraction of the minor component.

 $h = Part thickness (\mu m).$

 $I_{\rm C}$ = Layer breakup parameter (μ m).

The HDPE - Adhesive - EVOH system discussed previously can be used in combination with *equation 3* as an example to illustrate expected wall thickness effects in LIM. Initial layer breakup was observed in the EVOH minor component, present at 8 vol%, The onset of layer breakup occurred at an average EVOH layer thickness of approximately 0.3 µm in a 1 mm thick part. The EVOH component was present as the core layer in the feedblock. For these materials and compositions, the maximum permissible number of feedstream layers can be estimated, and selected values are tabulated below:

| h (mils) | M_p |
|----------|-------|
| 80 | 2100 |
| 40 | 1100 |
| 20 | 530 |
| 10 | 270 |

The limit of LIM technology will be reached when the minimum number of layers necessary to ensure adequate compositional uniformity exceeds the maximum permissible number of layers to ensure a sufficient degree of layer continuity.

<u>Multicavity Molding</u>: Recent research has used the previously described 16 cavity mold to assess the LIM process for multicavity injection molding. Preliminary results suggest substantial intracavity lamella retention. Figure 11 is an optical photomicrograph of a multicavity part cross-section taken from the region marked with an *x* in *figure 6*. The materials used were pigmented HIPS resins, and the parts were lamellar molded using a 129 layer feedstream. Substantial lamella retention was observed, even at the extremities of the mold cavity. One should note that the incident melt stream is sectioned into four quadrants prior to entering the runner system, and this results in the number of feedstream layers entering each runner to be reduced by one half (i.e., from 129 to 65 layers). The latter number approximates the number of layers visible in the total cross-section shown in *figure 11* (total part cross-section is not visible in the photo).

CONCLUSIONS

A lamellar injection molding process has been developed which allows the molder to create a layered morphology of two or more dissimilar polymers via direct molding, which can effect enhanced properties compared to conventional blend analogs. In addition to avoiding the cost of polymer compounding, compatibilizing polymers can be introduced at the interfaces between incompatible polymer pairs. Property enhancements which can be realized include barrier to gases and solvents, chemical and temperature resistance, dimensional stability and optical clarity. LIM can offer process simplicity and lower cost molds compared to coinjection molding, and allow for molding of complex multicavity parts. The process has been successfully scaled-up to a 715 ton reciprociating screw injection molding machine using standard molds. It is anticipated that commercial multicomponent injection molding machines can be easily adapted for LIM. Evaluation of the LIM process for specific applications is beginning.

ACKNOWLEDGMENTS

The authors would like to acknowledge numerous coworkers who have been instrumental to the successful implementation of LIM technology

Nothing herein grants or implies license to practice under patents of The Dow Chemical Company or others.

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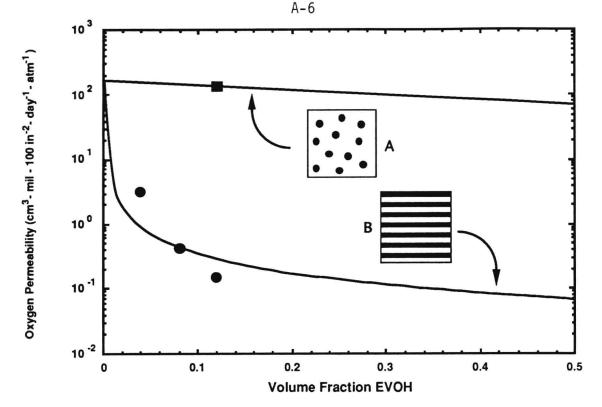


Figure 1. Oxygen permeability versus composition for a HDPE - Adhesive - EVOH material system which exhibits two morphological extremes. (a) Random dispersion, and (b) Perfect layer continuity. EVOH = Poly(ethylene-co-vinyl alcohol) (29 mole% ethylene).

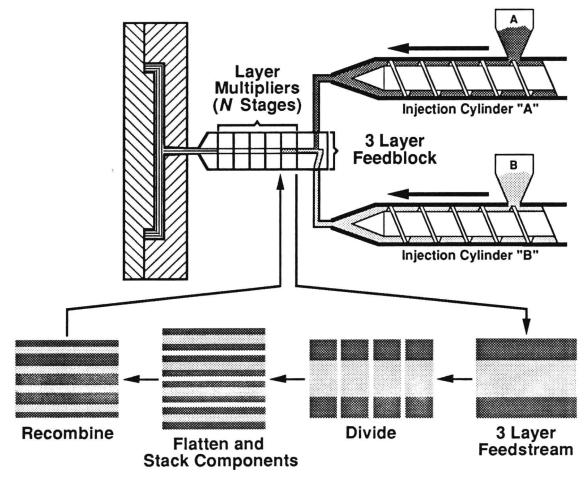


Figure 2. LIM process schematic.

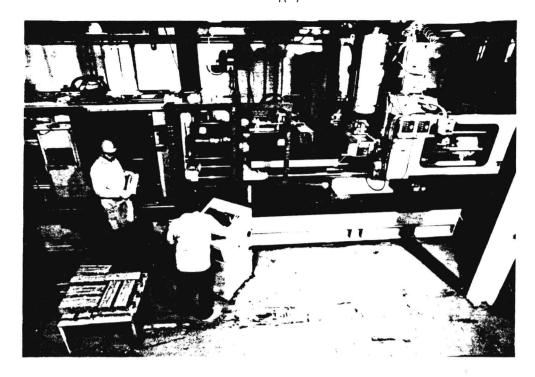


Figure 3. Pictorial illustration of the LIM process as practiced in developmental scale-up using a 715 ton reciprocating screw injection molding machine.

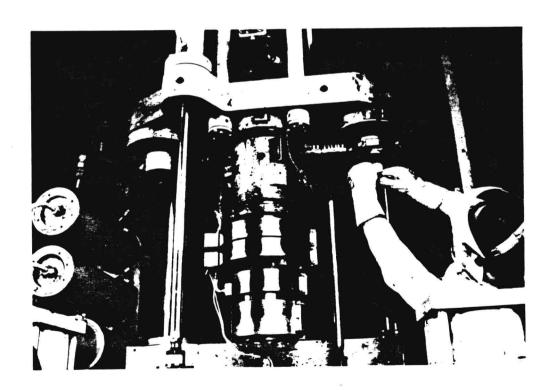
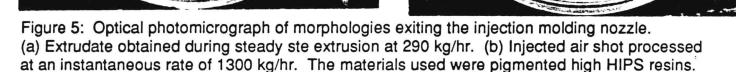


Figure 4. Exploded view of the feedblock and layer multiplication stages shown in figure 3.

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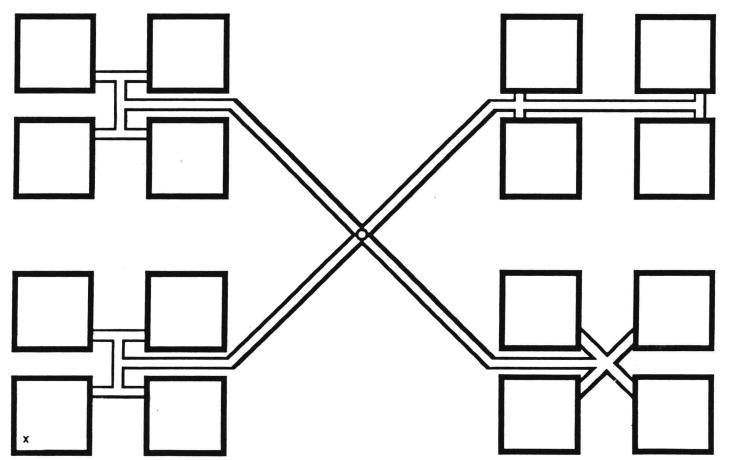
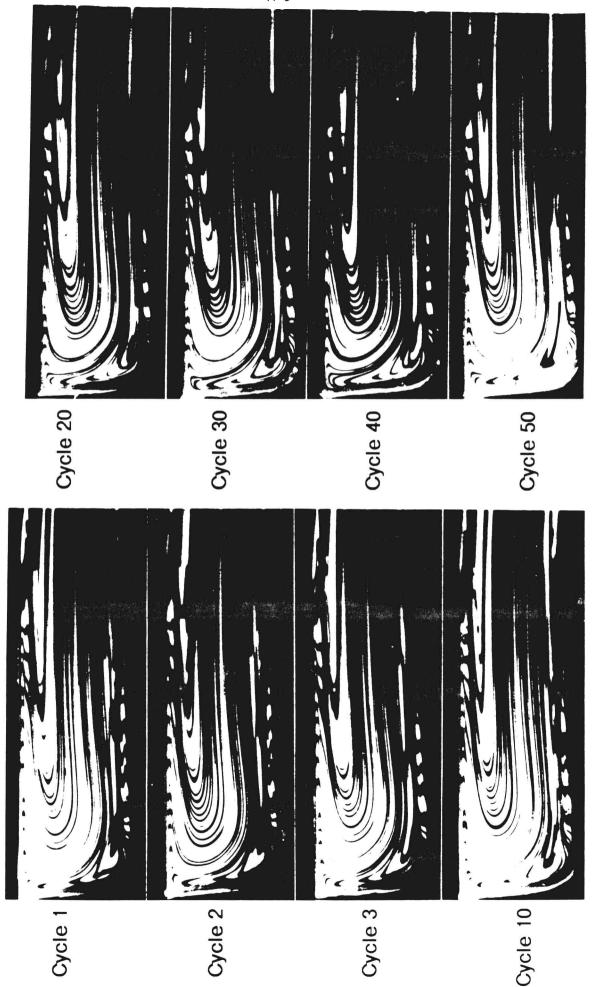


Figure 6: Schematic diagram of 16 cavity mold built to evaluate molding via LIM. The mold uses a trapezoidal runner system. Each individual mold cavity is 4" x 4" x 0.150" thick.



2 Em 2

cross-sectioned at identical locations from bumper beam moldings near the end of the flow path. The materials used Figure 7. Cross-sectional photomicrographs that illustrate the repeatability of the LIM process. Specimens were were high impact polystyrene pigmented blue and white to provide good optical contrast (70% white - 30% blue).

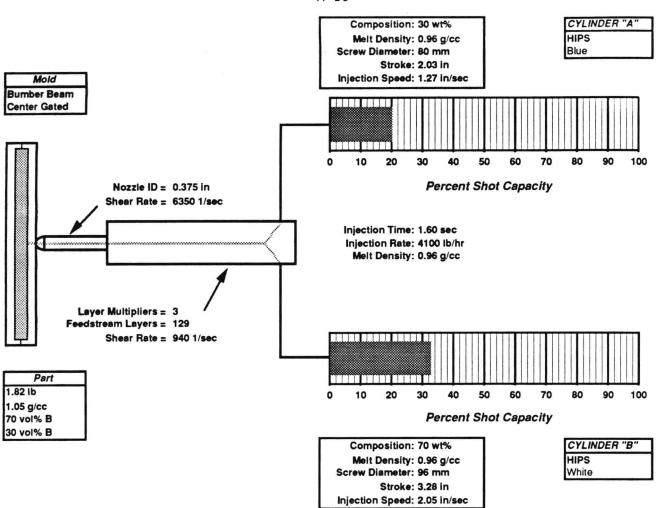


Figure 8. Injection molding conditions used for repeatability study.

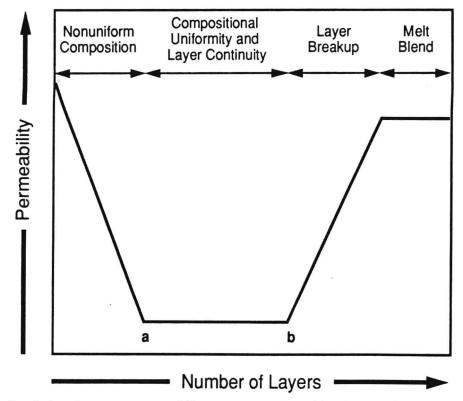


Figure 9. Idealized plot of oxygen permeability versus number of feedstream layers that illustrates the effect of layer thickness for a LIM multiphase barrier system of constant composition.

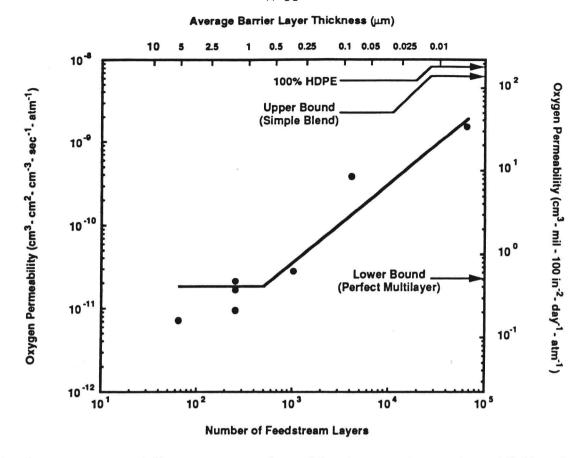


Figure 10. Oxygen permeability versus number of feedstream layers for a LIM barrier system comprised of HDPE-adhesive-EVOH (8 vol% EVOH).

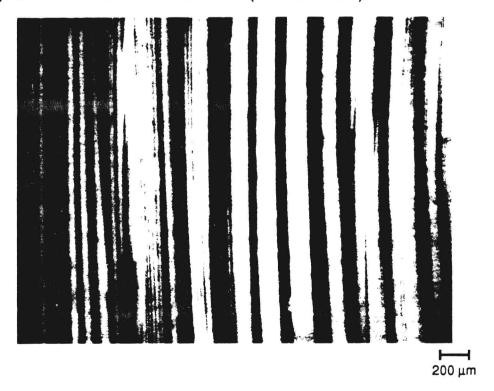


Figure 11: Molded part morphology resulting from initial multicavity molding trials using a 16 cavity tool. The illustrated part cross-section was taken from the marked region region in figure 6. Materials used were pigmented HIPS resins.

TITLE
EXPERIMENTAL CASE STUDIES OF DIMENSIONAL REPEATABILITY ON GAS ASSISTED
INJECTION MOLDING PROCESS AND CAVITY PRESSURE CONTROLLED CLOSED LOOP
INJECTION MOLDING FOR STRUCTURAL PARTS.

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ABSTRACT

The objective was to report the repeatability evaluations of cavity pressure feedback control injection molding vs. gas assisted injection molding for tight tolerance structural applications. It is the intent of this case study to compare the dimensional repeatability capabilities of these processes. This would assist engineers in determining which process to use for their specific applications. It was interesting to observe that cavity pressure feedback control on injection molding demonstrated better dimensional repeatability. The result also showed gas assisted injection molding to demonstrate better flatness repeatability.

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INTRODUCTION

It is the purpose of this work to report the dimensional repeatability evaluations of cavity pressure controlled injection molding and gas assisted injection molding processes for tight tolerance structural applications. It is widely acknowledged that gas assist molding yields stress-free parts, less warpage, and good dimensional control. In some people's minds, these attributes make this process ideal for tight tolerance applications. A tight tolerance process, however, must be repeatable, above all other attributes.

Various gas assist technologies have been theoretically and experimentally investigated to show the readiness of the process and understanding of the manufacturing and engineering capabilities^{1,2}. The advantages and disadvantages of the processes have also been reported³. No work has been reported with consideration of the dimensional repeatability of this process as compared to solid injection molding.

The repeatability (i.e.variations) of part dimensions in a gas assisted injection molding process need to be investigated. Understanding these variations and how they influence the final part dimensions is very important to design and process engineers in determining which process to use for their desired applications. An experimental case study on this topic was recently done in our laboratory and the results are shared in this paper.

EXPERIMENTAL METHODS

PROCESS

Cavity pressure controlled injection molding and the Cinpres IIR version of gas assisted injection molding processes were used in our investigations. All the molding trials were done at GE Plastics' Polymer Processing Development Center using a structural business equipment part (see figure IV). The first stage molding trials were conducted to set the baseline on repeatability with cavity pressure control. In order to achieve "apple to apple" comparison of these two processes, the same mold, with very minor modifications, the same molding machine, the same technician, and the same materials were utilized. All the moldings were done using the best known practices for the two processes. Since specific dimensions were not a criterion for quality, the part appearance and lack of sink and flash were used to determine processing parameters. During the molding trials, each of the processes were allowed to stabilize before fifty consecutive parts were collected for measurements.

MOLDS

A P-20 production steel tool for a business equipment part approximately 406.4mm x 63.5mm x 89mm with a nominal 3.2mm wall section was modified for cavity pressure controlled injection and gas assited injection molding trials respectively. For the cavity pressure controlled moldings, the locating ring was modified to fit GE Plastics molding machine. A center sprue fed a runner system with four gates symmetrically placed within a center opening in the part. A Dynisco cavity pressure transducer was added under an ejector pin which was located approximately halfway between the gate and end of flow to monitor the cavity pressure, and control the molding machine.