Visualization of Bubble Dynamics in Foam Injection Molding

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ABSTRACT: This article presents a visualization study on nonisothermal bubble growth and collapse in the foam injection molding process (FIM). Observation study can give more insight to the bubble growth in foaming process, especially in the challenging injection foaming process. In this study, besides the growth of bubbles, collapse of the bubbles was also observed which could provide knowledge to the final foam morphology. Cell growth vs. time was recorded and analyzed using a software-equipped high speed camera. To investigate the cell collapse, various holding pressure was exerted on the gas-charged molten polymer. The amount of holding pressure had noticeable effect on the rate of bubble collapse. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 116: 3346–3355, 2010

Key words: foam injection molding (FIM); visualization; cell growth; cell collapse; critical bubble radius

INTRODUCTION

Polymeric foams are plastic processed materials with great usages and commercial aspects. This gives a great motivation to many researchers to study the foaming behavior both theoretically and experimentally.¹⁻⁹ Among all the foaming processes, foam injection molding has greatly been under consideration because of its challenging nature.

Foam injection molding includes four major steps: (1) dissolution of a specific amount of blowing agent (chemical or physical) into a molten polymer, (2) nucleation of tiny bubbles in gas-charged molten polymer, (3) growth of the nucleated bubbles, and (4) formation of a molded part with cellular structure. Growth stage in foaming process involves diffusion of gas into the nucleated sites by means of lowering the system pressure. Among all, cell growth phenomenon has been studied by various scientists and researchers. In addition, investigation of nonisothermal bubble growth can be more useful as it represents the real situation. The role of heat transfer on growth and collapse of bubbles is dominant as the majority of gas-charged molten polymer properties such as solubility, diffusion, Henry's constant, melt compressibility are function of temperature. A large body of literature exists on the subject

of bubble kinetics in Newtonian or viscoelastic liquid medium. The first work presented by Barlow and Langlois¹ considered the growth of a stationary single bubble in a purely viscous (Newtonian) liquid and isothermal condition, as well as taking into account both the diffusion and momentum transfer process in their theoretical investigation. Street² carried out a theoretical study on bubble growth in a viscoelastic medium by considering the momentum transfer and neglecting diffusion phenomenon between liquid and gas phase. He found that bubble growth is faster at the early stages in viscoelastic liquid comparing with the corresponding Newtonian liquid with the same shear viscosity. Later, Street et al.³ studied free expansion bubble growth by considering finite influence volume around each bubble in a viscous, non-Newtonian (power law) liquid and nonisothermal condition. Their study considered momentum, mass and heat transfer processes. Villamizar and Han⁴ carried out a visual experiment on bubble dynamics in structural foam injection molding using a rectangular mold cavity and showed that bubble dynamics during mold filling is noticeably affected by injection rate (or injection pressure), melt and mold temperature, viscoelastic properties of molten polymer, blowing agent concentration and gas dissolution and diffusion into or out of molten polymer. In their study, four-fifths of the cavity was filled by short-shot injection and then filling was completed by the expansion of the gascharged molten polymer. They also studied bubble collapse during mold packing in structural foam processing in isothermal state. Han and Yoo⁵ carried

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out an experimental and theoretical study on bubble formation and growth in isothermal condition at fast injection rates including diffusion process, interfacial tension and stress relaxation of polymeric melt. They concluded that at high injection pressure, bubble growth will be retarded in the early stage of foam injection molding. They also filled four-fifth of the cavity and then growth of the bubbles filled the rest of the cavity. Amon and Denson⁶ presented a mathematical analysis of bubble growth in expanding foam based on cell model in viscoelastic medium and isothermal condition. Arefmanesh et al.⁷ utilized the unit cell model to a low pressure structural foam molding to study simultaneous growth of a given number of bubbles assuming a Newtonian medium. They continued their researches and mathematically studied diffusion induced bubble growth in viscoelastic medium by considering upper-convected Maxwell model to describe rheology of the fluid.⁸ Later, they developed a numerical method to model simultaneous growth of a large number of bubbles under nonisothermal condition and low pressure during foam injection molding.9 They used a hydraulic cylinder to inject gas-charged molten polymer into the mold. Using this method, cell nucleation could be initiated before and during the injection stage of the process completely. Also, due to a lack of mechanism for mixing polymer and the blowing agent during the melting stage, excessive coalescence was occurred between the cells. Yokoi et al.¹⁰ developed a two axis tracking system and measured the position of flow front dynamically. They also observed some typical injection molding defects (e.g., silver streaks and gas bubbles generation) in their study. Later, Han and Yokoi¹¹ studied the behavior of a polymeric melt flowing over a stamper with V-grooves during filling stage of injection molding using a prism glass-inserted mold. Yamada et al.12 studied the filling stage of microcellular foam injection molding using glass inserted visual mold and investigated the formation mechanism of layers during foaming process.

Osorio and Turng¹³ developed a mathematical formulation and numerical simulation for nonisothermal bubble growth during postfilling stage of microcellular foam injection molding. In this study, they solved the equation of mass diffusion within the envelope rather than assuming a polynomial profile. Chen et al.¹⁴ studied plasticization effects on bubble growth during polymer foaming. Behravesh and Rajabpour¹⁵ carried out an experimental investigation on filling stage of microcellular foam injection molding with main emphasize on the cell growth stage via variation of shot size and concluded that formation and growth of cells strongly depend on shot size. In addition, they found that a high shot size causes formation of single phase solution of gas/polymer system. They also presented a foaming characteristics diagram for microcellular foaming process. Yue et al.¹⁶ developed an arbitrary Lagrangian-Eulerian scheme to track bubble surface during foaming process as well as simulating diffusion induced growth of a 2D foam and thinning of a film between a growing bubble and free surface. Xu et al.¹⁷ studied the growth and collapse behavior of chemical blowing agent (CBA)-based foaming process under atmospheric pressure using a hot-stage optical microscopy digital imaging setup. They also proposed a model that included diffusion, surface tension, viscosity, and elasticity effects. They concluded that temperature, gas bulk concentration and diffusivity play significant roles on CBA-blown bubble life span. Leung et al.¹⁸ studied the change in the critical bubble radius and its effect on cell stability during foaming process. In their study, bubble growth and collapse phenomena were theoretically investigated in a CBA-based, pressure free foaming process. They also performed some experimental investigation on bubble growth and collapse to verify their model using hot-stage optical microscopebased image processing system.

The exploration of literatures indicates that although a large amount of work have been attended toward bubble growth dynamics, less attention has been given to the online observation of the bubble growth process in nonisothermal state and also collapse stage. This work specifically concentrates on the observation of cell growth and collapse under a three stage sequence of "injection, holding and pressure removal" in nonisothermal conditions. Also, filling the cavity (full shot injection) could limit the number of bubbles created, and hence, they are easier to track. In other words, in short shot, because of a larger room for expansion, too many bubbles are nucleated, grown, and merged (cells coalescence) so that the study on a single bubble would be highly difficult. This could ease the visualization of the collapse phenomenon and its relation to the system pressure.¹⁵

EXPERIMENTATION

Mold design

Figure 1 shows a schematic of the mold used in this study. A detailed description of the designed mold can be found in the references.^{19,20} A rectangular cavity ($62 \times 50 \times 5.5$ mm) was manufactured with the specification shown in Figure 2. To retard cooling of the sprue, new sprue was designed and manufactured with a larger inner conic hole compared with the standard available sprues.

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View Direction



Figure 1 A schematic of the visual mold.

Equipment and materials

Figure 3 shows a schematic view of prepared setup consisting of a 70-ton injection molding machine, CO_2 gas injection apparatus, a high speed camera (FASTCAM-super 10K-Photron Ltd.), light source, motion analyzer, a computer, and the visual mold. Gas injection system was prepared consisting of a CO_2 gas cylinder, a dosing valve (micro-valve), an on-off valve and a non-return valve to inject specific amount of CO_2 gas in to the melt. Polystyrene, grade TAITAREX GPPS 861N, manufactured by Taita Chemical Co, was used as the polymeric material to produce the plate with specification depicted in Figure 2. This engineering thermoplastic has a good transparency, which makes it suitable for visualizing the bubbles formation.

Procedure

An injection molding machine was set and prepared to inject a specific amount of polystyrene melt into the mold cavity. Because of the high solubility of CO_2 in polymeric materials, 0.5 wt % of CO_2 gas was injected into the melt as a physical blowing agent (PBA) using a dosing valve, after the dosing system was calibrated. This little amount of PBA presents a condition to have a better control on the number of grown bubbles. A screw with a mixing section was used to fully disperse the CO_2 gas into the PS melt. To have a single phase solution in the barrel, the pressure of the PS-PBA system (back

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pressure) was adjusted above the solubility pressure. Melt injection temperature and mold wall temperature was set at 290°C and 15°C, respectively. Mold cavity was filled completely by a full shot. To study the collapse behavior of bubbles, holding pressure was applied to pack the gas-charged molten polymer. Formation, growth stage, and collapse of bubbles were then recorded using a high speed camera with recording speed of 30 frames per second. Captured pictures were then analyzed using image processing software to extract the required information of foaming behavior at each desired time interval. The visualizing apparatus in this work could clearly detect bubbles with 400 micron radius. A higher magnification, for detecting the smaller size, could limit the visualizing area, which in turn, could make tracking of a moving object (here a nucleated cell) in an unpredictable path highly difficult or even impossible with current system. Hence, a $3 \times$ magnification lens was used to view whole area of the cavity under study. For all experiments, PS melt was injected into the cavity with 15 MPa injection pressure. Holding pressure was varied via a control pressure valve of the machine.



Figure 2 Specification of the part and designed sprue.

Figure 3 Schematic view of the prepared setup.

RESULTS AND DISCUSSION

Rheological properties of gas-charged molten polymers are noticeably affected by concentration of PBA. In addition, injection pressure is an important parameter which strictly affects formation and growth of bubbles in molten polymer.⁵ Figure 4 shows photographs taken during filling stage of PS/ CO₂ melt system in the cavity with an initial melt temperature of 290°C. Full scale view of the mold cavity, sprue and gasket can be seen in the first frame. Before injecting the polymeric melt into the cavity, PS-CO₂ system was pressurized in the barrel to ensure a single phase solution. As the nozzle opens and melt enters into the mold cavity, because of the pressure drop down below the solubility pressure, myriads of bubbles nucleate and grow continuously in the molten polymer and form a milky color melt. Growing bubbles move with the motion of flow front. Because of the pressure gradient in the mold cavity, the velocity of the nucleated bubbles is greater than that of flow front and this causes some bubbles to reach flow front and burst eventually. To demonstrate this, an experiment was performed to clearly show the difference between the velocity of the gas bubbles and the velocity of the flow front. For this purpose, a condition was prepared so that a few bubbles are formed inside the molten polymer moving along with the flow front. Another screw with shorter mixing section was used and the gas blowing agent amount was decreased (via regulating the micro valve on the lowest level possible). Figure 5 shows some photographs taken during the performed experiment.

As the cavity was fully filled by the gas-charged molten polymer, the pressure inside the cavity was increased (via maintaining the holding pressure), and then consequently, most of the bubbles were suddenly collapsed [Fig. 4(i)]. This can be observed in the three last frame images of Figure 4. Interestingly, some are not collapsed completely and survive in the system. The remained bubbles are most located at the second half of the cavity. Pressure gradient (because of pressure loss in the mold cavity), temperature gradient, and blowing agent concentration gradient could cause this difference. Lower pressure at the flow front could promote higher cell nucleation resulting in more and larger bubble presents at the front region. Besides, as illustrated in

Figure 4 Visualization of cell growth and collapse during filling stage. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Figure 5 gas bubbles tends to move faster than the adjacent melt resulting in commutation of more bubble at the end region.

Note that the last frame image shows some bubbles burst on the surface of the foamed plate and forms silver strikes patterns [Fig. 4(j)].

Visualization of cell growth in FIM

Bubble dynamics (bubble growth and/or collapse) during mold filling depends on many factors. One

important is injection pressure, which dominantly determines how soon the bubbles will nucleate and grow.⁵ In the present experiments, cavity was filled via a full shot. After filling the cavity by the gaspolymer system, it took some time to initiate bubble growth and subsequently reaching the visualization limit. Figure 6 gives a better understanding of cell growth steps in a non-isothermal condition. As it can be seen in the images, gas diffuses into the bubbles and gradually expands the bubbles up to a limit where the gas is depleted from the surrounding

Figure 5 Photographs demonstrating the greater velocity of gas bubbles compared with flow front velocity. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

matrix or too high an increased melt viscosity opposes the growth. It should be pointed out that grown bubbles have different sizes. The size difference could be due to pressure and temperature gradient in the system caused by heat transfer of the mold walls. As no nucleating agent was used in our experiments, few bubbles tend to grow. Another important factor which decreases the number of grown bubbles is the system pressure (or holding pressure). Regardless of the nucleation mechanism, increased pressure in the mold cavity increases the size of the critical radius according to the classical nucleation theory:

$$R_c = \frac{2\sigma}{P_{\rm bub} - P_{\rm sys}} \tag{1}$$

$$\Delta G_{\rm hom}^* = \frac{16\pi(\sigma)^3}{3(P_{\rm bub} - P_{\rm sys})^2}$$
(2)

$$\Delta G_{\text{het}}^* = \frac{16\pi(\sigma)^3}{3(P_{\text{bub}} - P_{\text{sys}})^2} S(\theta)$$
(3)

where σ is surface tension, P_{bub} and P_{sys} are gas pressure inside the bubble and system pressure, respectively, and $S(\theta)$ is a function of θ , wetting angle.²¹ Therefore, bubbles with radius above the critical radius in the system will continue to grow, and the growth of bubbles with the size smaller than R_c will be ceased. This means that a smaller number of bubbles could have chance to nucleate and grow in the system (with high pressure) compared with the situation of a free expansion. Cell coalescence, which in most of the time occurs because of the pressure difference between two neighboring bubbles, can be noticed in the last frame images of Figure 6. To sketch bubble radius versus time, frame images were analyzed using image processing software (microstructure measurement produced by Nahamin Pardazan Asia Co). The error occurred in image processing procedure was less than 5%.

For each experiment, one target cell was chosen having an approximately spherical shape and medium in size compared with the other present cells grown in gas-charged molten polymer. Figure 7 gives a plot of bubble radius versus time for a target cell indicated in Figure 6(d). It should be mentioned that bubbles are more spherical at the beginning of growth stage. Then some gradually tend to deform to a nonspherical shape [Fig. 6(g)]. The main reason for this issue is the removal of holding pressure, which is conventionally applied to suppress excessive shrinkage. Areas near the gate shows lower shrinkage compared with the areas at the end of the flow. But interestingly, if the packing pressure is not maintained until the gate or runner is frozen, then the pressure in the cavity will cause the melt to flow back into the runner system. This can result in a higher shrinkage around the gate area than the rest of the cavity. Thus, an uneven shrinkage occurs in

Figure 6 Visualization of nonisothermal bubble growth in FIM. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

the cavity and consequently bubbles tend to deform to a nonspherical shape. This phenomenon can be observed more clearly for the larger cells.

Visualization of cell collapse in FIM

Figure 8 shows photographs of cell collapse steps resulted from exerting 10 MPa pressure on the gascharged molten polymer. Pressure was exerted to the system similar to the holding stage in a conventional injection molding using the ram action of the screw. Holding pressure was also exerted for 25 s. First frame image shows the end step of the growing stage at t = 20.1 s. The second photograph [Fig. 8(b)] reveals that bubbles located at the middle of the foamed specimen are collapsing, while those at the end of the cavity are not yet affected by the pressure. However, continuing the exerting pressure leads to collapse of more bubbles. The last frame image of Figure 8 shows that only few bubbles are not collapsed completely. Two factors can justify this phenomenon: (1) increase of the melt viscosity of the

gas-charged molten polymer and (2) not a high enough exerted pressure to cause the remained bubbles to collapse.

Similar experiments were performed with two different holding pressures of 15 and 18 MPa. Interestingly, these pressures were high enough to

Figure 7 Graph of bubble radius versus time for the target cell.

Figure 8 Cell collapses under 10 MPa pressure. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

overcome high viscosity (caused by decreasing temperature in the foamed specimen) and to completely collapse the target cells. Figure 9 gives the plots of cell collapse under three different pressures. It should be mentioned that at holding pressures of 15 and 18 MPa, although the target cell size decreased under the visualization limit (400 micron) to be measured quantitatively, but still the cell could be detected, and thus, located to observe the re-growth. As it can be seen in this figure, increasing pressure affects the slope of the R-t curve so that the higher the holding pressure, the steeper the slope of the R-t curve. Similar observation was reported by Villamizar and Han⁴ in an isothermal condition. But it should be mentioned that in a nonisothermal condition, because of the increase in viscosity during the time, the rate of bubble collapse is lower than that of the isothermal condition.

Equation (1) coupled with Figure 10 best describe this phenomenon. Consider a bubble with radius R_b grown in the system. Each system, depending on its pressure, P_{sys} (here, holding pressure), has a defined critical radius, R_c . In this case, target cell in the gascharged molten polymer could resist a complete collapse under a holding pressure of 10 MPa, whereas target cells with the same radius could collapse completely under 15 and 18 MPa holding pressure. In other words, the collapse phenomenon can be explained by the classical nucleation theory; when the system pressure is low, the critical bubble size is low and then a major number of bubbles tend to

Figure 9 Collapse of target cells under different holding pressure. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Figure 10 The excess free energy of bubbles with different system pressure.

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0000 0 10 MPa (holding pressure) 0 15 MPa 18 MPa 1000 1000 1000 0 5 10 15 20 25 30 35 40 45 50 55 60 65 70 75 Time (s)

Figure 11 Bubble radius vs. time for three stages: filling the cavity, under holding pressure (for 25 s), and releasing pressure.

grow. When the system pressure increases, the critical bubble size increases, and then those bubbles which have the size smaller than this critical size will tend to collapse. Increasing the pressure of system increases the critical bubble size, and then more bubbles tend to collapse until no bubble will survive in the system. The gas inside the bubbles diffuses back into the polymer matrix and dissolves in it as it can be verified in the reference.¹²

 $R_c(10 \text{ MPa}) < R_b < R_c(15 \text{ MPa})$

Figure 11 shows a complete history of bubble dynamic (growth, collapse, and then growth) obtained in three different system (holding) pressures of 10, 15, and 18 MPa. It contains the third stage where the holding pressure is then removed to observe the bubble growth dynamics. This figure reveals that higher holding pressure retards growth of collapsed bubbles after removing the holding pressure. At a higher holding pressure, the system pressure is higher and more time is needed to reduce the pressure to a limit at which the bubbles can grow again in the system. It should be mentioned that for all the experiments holding pressure was exerted to the gas-charged molten polymer for 25 s (after which the pressure was removed to allow bubble growth). However, as Figure 11 reveals, slope of the R-t diagram approaches to zero after bubbles re-grow within few seconds.

Figure 12 shows growth of the collapsed bubbles after removing the holding pressure. It is seen that bubbles have not reached their previous sizes because of the nonisothermal state of the system and so of the increasing viscosity of the polymeric melt. However, this system has its own critical radius and so few bubbles could grow again in the gas-charged molten polymer. This could suggest a method to control bubble growth, and thus, their size in foam injection molding.

CONCLUSIONS

This study presents visualization of the cell growth and collapse phenomenon under a nonisothermal condition. While the majority of the former studies focused on the investigation of bubble growth under

Figure 12 Growth stage of the collapsed bubbles after removing holding pressure (10 MPa). [Color figure can be viewed in the online issue, which is available at www. interscience.wiley.com.]

a free expansion situation, this work examined the bubble growth under a pressurized system. Photographs obtained by the prepared setup showed that bubbles, created by dropping pressure below the solubility pressure, moved with the flow front. The bubbles tend to collapse when the pressure of system is increased (holding pressure). This phenomenon can be explained by the classical nucleation theory. Visualization of bubble growth was also carried out after pressure removal. The history of bubble growth was given and it showed that holding pressure has a dominant effect on the dynamics of bubble growth. Especially, it was shown that the holding pressure plays a dominant role on the second growth stage and also final cell size. This could give insight to the better control of foam injection molding process.

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