Bubble Growth in a Viscous Liquid in a Simple Shear Flow

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The nucleation and growth of a bubble in a Newtonian liquid in simple shear and creeping flow were studied experimentally in a Couette apparatus operating under vacuum. A low molecular weight polyisobutylene was chosen as a typical high-viscosity Newtonian liquid. The volatile solvent in our experiments was simulated by air, and the formed or growing bubbles were photographed under shear. The experimental foaming results indicate that vacuum (at a level of supersaturation conditions) is not sufficient to boil a viscous solution, even in the presence of bubbles (nucleation sites), as shear also has to be applied. Furthermore, there is probably a critical shear rate (or stress) at which boiling appears. Bubble growth experimental results indicate that the growth rate of a slender bubble increases as the shear rate increases and as the pressure decreases. The experimental results agree well with theoretical models developed by Lee and Biesenberger (for bubble nucleation) and by the authors (for bubble growth).

Introduction

In the polymer processing industry, mass transfer between a bubble and a viscous liquid can be found in the devolatilization step, where low concentrations of volatile components are removed from the polymer melt. This process, which is conducted at high temperatures and low pressures through a boiling−foaming mechanism, is accompanied by the formation and growth of bubbles. A recent book on the subject of polymer devolatilization edited by Albalak (1996) has been published.

In general, formed or growing bubbles cannot always be considered as spheres or even slightly perturbed spheres. This is especially true when bubbles are present in high-viscosity liquids such as polymer melts, processed in rotary machines in which high shear rates can be developed. In this case, elongated bubbles with pointed ends are observed. An excellent review on the subject of bubble deformation in viscous shear flows was given by Rallison (1984).

There is a large number of works in the literature dealing with the nucleation or growth of a spherical bubble in a quiescent liquid. Very little is known about those two phenomena in the presence of a shear field. Yet, experimental results together with theoretical considerations of the very few articles published on the subject, suggest that shear fields can enhance the formation and growth of bubbles (Lee, 1996; Favelukis et al., 1995).

In our previous works we studied the deformation (Canedo et al., 1993) and the dissolution (Favelukis et al., 1995) of a slender bubble in a Newtonian liquid in simple shear and creeping flow. The present article, which is a continuation of the previous works, deals experimentally with the nucleation and growth of a slender bubble in simple shear flow.

Theory

Bubble nucleation

Lee and Biesenberger (1989) reviewed the different theories for bubble nucleation. With their previous experimental

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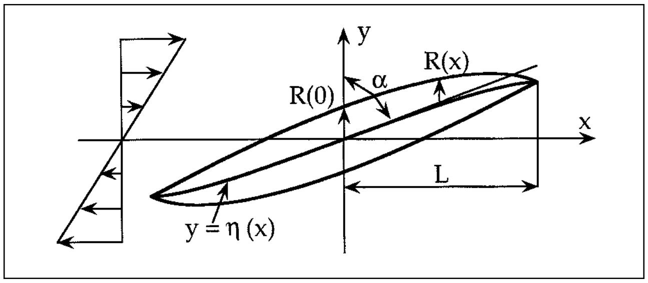

Figure 1. Slender bubble in a simple shear flow.

$R(x)$ is the local radius, L is the half-length of the bubble, α is the inclination angle, and η is the bubble's center line.

results (Biesenberger and Lee, 1985, 1986a, 1986b, 1987), they concluded that the most suitable theory that explains boiling in a devolatilization process is the one that assumes heterogeneous nucleation from a microscopic cavity in which gas is entrapped. This type of nucleation site can be formed as a result of holes, scratches, and so forth, which are always present on the walls. Alternatively, the site can appear inside the liquid in the form of a contaminant (for example, inside a dust particle). This type of site was suggested many years ago by Harvey et al. (1944), and it can also explain the beginning of boiling in regular pure liquids.

Lee and Biesenberger performed a small modification in Harvey's model, and they called it the metastable nucleation site (see Biesenberger and Lee, 1986b; Lee, 1991, 1993, 1996). According to the model, a force balance shows that in polymer melts, the only dominant acting forces are shear forces (which tend to "pull") and surface tension forces (which tend to "keep" the gas phase inside the nucleation site). Only by deformation in the melt, in addition to the presence of shear forces, which can overcome the surface tension forces, is it possible to extract part of the entrapped gas within the cavity to form a free bubble. Therefore, conditions of supersaturation alone are not enough to boil a polymer solution; shear is also needed.

Bubble deformation

Figure 1 describes a slender bubble (inviscid drop) with a local radius $R(x)$ and a half-length, L, placed in a simple shear flow. The velocity components of the undisturbed motion are given by

$$v_x = Gy \tag{1}$$

$$v_y = 0 \tag{2}$$

$$v_z = 0 \tag{3}$$

where G is the shear rate.

An approximate solution to the deformation problem in creeping flow has been presented by Hinch and Acrivos (1980), under the assumption that the cross section of the bubble is circular. The following dimensionless parameters can be obtained (Canedo et al., 1993):

$$R^*(0) = \frac{R(0)}{a} = 0.578\,Ca^{-1/4} \tag{4}$$

$$L^* = \frac{L}{a} = 3.45\,Ca^{1/2} \tag{5}$$

$$A^* = \frac{A}{4\pi a^2} = 1.41\,Ca^{1/4} \tag{6}$$

where a is the radius of a sphere of equal volume, A is the surface area, and Ca is the capillary number defined by

$$Ca = \frac{\mu G a}{\sigma}, \tag{7}$$

where μ is the viscosity of the liquid and σ is the surface tension. Note that in order to obtain a slender bubble ($R/L \ll 1$), the condition $Ca^{3/4} \gg 1$ must be met.

The theoretical solution obtained by Hinch and Acrivos (1980) suggests an S-Shaped bubble. The bubble becomes thinner, longer, its surface area increases, and it forms a larger angle with the y-axis with an increase in the capillary number. This theory was experimentally checked in our previous work (Canedo et al., 1993) in a Couette apparatus using air bubbles in low molecular weight polyisobutylenes. The experiments confirmed the theoretical model; however, it was discovered that the cross-section of the bubble is not circular.

Bubble growth

Assuming the thin concentration boundary-layer approximation, we presented (Favelukis et al., 1995) a simple (order-of-magnitude) model for the steady mass transfer of the volatile solvent to or from a nonbuoyant slender bubble in a simple shear and creeping flow. Applying a quasi-steady-state approach, an expression for the rate of growth (or dissolution) has been suggested for the case in which the gas inside the bubble contains only the volatile component, which behaves as an ideal gas of constant pressure and temperature. The following result has been obtained:

$$\frac{da}{dt} \sim \frac{R_g T (c_\infty - c_s)}{P} D^{1/2} \left(\frac{\mu}{\sigma}\right)^{-1/8} a^{-1/8} G^{3/8}, \tag{8}$$

where R_g is the universal gas constant, T is the absolute temperature in the bubble, and P is the pressure inside the bubble. The molar concentrations of the volatile solvent at the bubble surface (c_s) and far away from the bubble (c_∞) were assumed to be uniform and constant. Finally, D is the diffusion coefficient and t is the time.

The most important result of the simple model is that the growth rate of the bubble depends on the shear rate to the power of 3/8. Thus an increase in the shear rate will increase the rate of mass transfer. This is an important result for the design of devolatilizers such as vented extruders, since it suggests higher mass-transfer rates at higher rotational speeds.

Equation 8 was developed and experimentally checked in our previous work (Favelukis et al., 1995). Air and carbon dioxide bubbles were dissolved in low molecular-weight polyisobutylenes in a Couette apparatus. No "tip streaming" was observed, and the experimental results compared favorably with the predictions of the simple model. Substituting Eq. 5 into Eq. 8, the expression for the growth rate can be written in terms of the half-length of the bubble:

$$\frac{dL}{dt} \sim \frac{R_g T(c_\infty - c_s)}{P} D^{1/2} \left(\frac{\mu}{\sigma}\right)^{1/4} L^{1/4} G^{3/4}. \qquad (9)$$

This last equation will be compared here with the experimental results. Note that as the bubble rises (due to the buoyancy force), the pressure decreases as does the solvent concentration at the bubble surface (assuming that Henry's law is valid). Then, according to Eq. 9, both phenomena cause the growth rate to increase.

In practice there are two more mechanisms for bubble growth. The first one is based on mass transfer as a result of the upward movement of the slender bubble. This type of growth is probably negligible since the rise velocity is very small, and since Eq. 8 predicted our previous experimental results of bubble dissolution. The second mechanism is bubble growth without mass transfer as a result of the decrease in the hydrostatic pressure as the bubble rises. Assuming an ideal gas in the bubble at constant temperature (PV = const.), then the contribution of the last mechanism to the total growth rate can easily be calculated. This contribution was found to be negligible compared to the experimental growth rates.

Experimental Results and Discussion

Using the experimental experience from our previous deformation and dissolution experiments, an improved version of a Couette apparatus was built. It is similar to the classic design, but it also can be covered in order to be operated under vacuum. Low molecular-weight polyisobutylene (PIB950, Exxon) was chosen as a typical high-viscosity Newtonian liquid. The volatile solvent in our experiments was simulated by air. More information on the experimental system and technique can be found in Favelukis (1995).

Since at atmospheric pressure the viscous liquid was already saturated with air, any reduction in the ambient pressure would have immediately caused supersaturation conditions. The use of air has another advantage, since contrary to a real solvent, it cannot dissolve in large quantities. Therefore, the viscosity and other physical properties of the liquid during the experiment are practically constant, allowing us the use of the bubble deformation correlations. All the photographs in this report were taken from the *x*-*z* plane (see Figure 1), where the S-shape of the bubble cannot be observed. The experiments were also recorded by a video camera.

Foaming experiments

At the beginning of an experiment, a vacuum was applied to the system, with the cylinders at rest until the absolute pressure over the liquid reached a value of 22 mmHg. The system was kept at this level for about 17 minutes. During this period of time no foaming was observed (see Figure 2a). However, a few bubbles appeared at the other side of the cylinders and therefore are not shown.

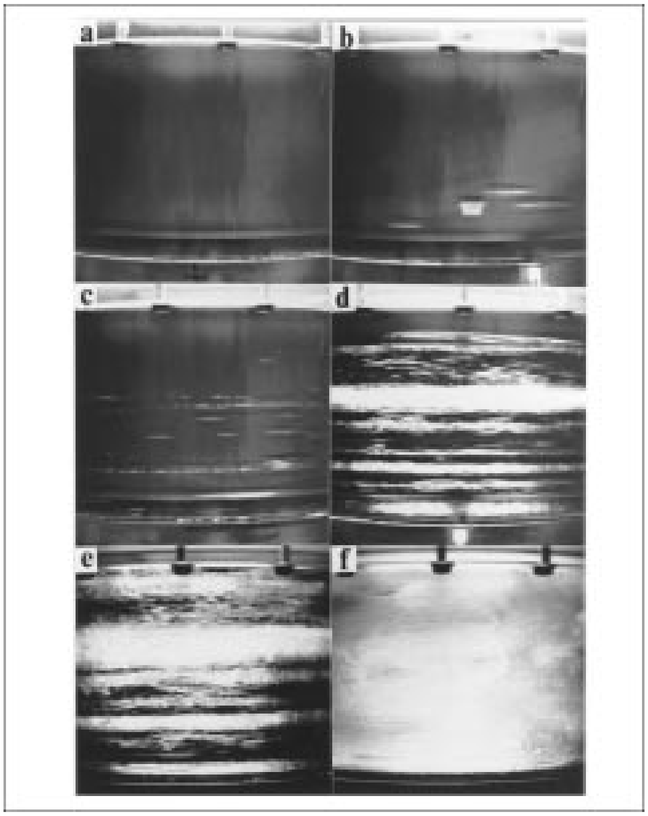

Figure 2. Foaming at 22 mm Hg.

(a) $G = 0$; (b) $G = 2.7$ 1/s; (c)–(f) $G = 12$ 1/s. In all figures the system is under vacuum.

Then, a low shear rate of 2.7 1/s was applied to the system while the two cylinders rotated at about the same velocity (but in opposite directions). This stage is shown in Figure 2b, where one can see a few elongated bubbles. These bubbles seem to grow due to the decrease in the hydrostatic pressure as they rise, and perhaps as a consequence of mass transfer. Foaming was not observed, and the system was kept at this lower shear rate for about 6 min.

At this point a high shear rate of 12 1/s was applied, and in a few moments boiling seemed to start (Figure 2c). The continuation of this fast process is shown in Figure 2d and 2e, and finally in Figure 2f, where the liquid is completely foamed. It took about 2 min from the beginning of the boiling process until one reaches the stage shown in Figure 2f. After 4.5 min of rotating the system at the higher shear rate, the motion of the cylinders was stopped while maintaining vacuum.

Figure 3a shows the cylinders at rest. The pressure is 23 mmHg. After approximately 6 min, the pressure was raised to one atmosphere, where the system was kept for about 7 min with no significant changes. At this stage, which is seen in Figure 3b, the liquid is again completely foamed, but the picture seems to be darker than in Figure 3a. This can be easily explained by the reduction in the bubble's volume as a result of the pressure increase.

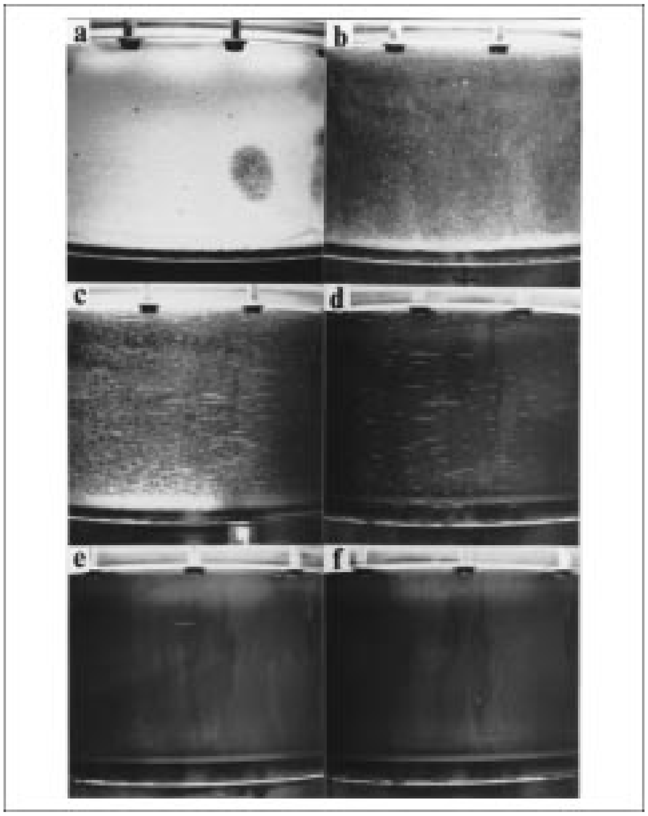

Figure 3. Dissolution at atmospheric pressure.

(a) $G = 0$, $P_0 = 23$ mm Hg; (b) $G = 0$; (c)–(e)$G = 12$ 1/s; (f) $G = 0$. In (b)–(f) the pressure is atmospheric.

Because of the high viscosity of the liquid it is clear that it would take a long time for the bubbles to reach the surface level or to dissolve. Therefore, and in order to maintain the air dissolved in the liquid for the next experiment, shear was applied to enhance dissolution. The cylinders were rotated at the high shear rate of 12 1/s and at atmospheric pressure. See Figure 3c, where again one can see elongated bubbles. The continuation of the dissolution process is shown in Figure 3d and 3e. In this dissolution experiment (at atmospheric pressure) the rotation was stopped after about half an hour.

Finally, the system at rest and atmospheric pressure are seen in Figure 3f, where the liquid is clear and transparent again and contains almost no bubbles. The temperature of the experiment shown in Figures 2 and 3 was in the range of 20°C at the beginning of the experiment to 23°C at the end. The rise in the temperature can be explained by viscous dissipation.

Another interesting observation was that after a series of experiments with the same viscous liquid and without emptying the system, foaming has not been observed both at low and high shear rates. We can imagine that with time the air content inside a nucleation site is reduced. Air can be very stable inside a nucleation site at subcooled conditions (for example, by forming a negative curvature radius inside a conical cavity). Once it exits the nucleation site, however, it cannot return. The result is that the polymer remains saturated with air, but with less air inside the nucleation site, since bubble growth was also observed. Furthermore, the introduction of air bubbles into the liquid at this point did not cause the system to foam at the low shear rate, while at the higher shear rate the system foamed. Similar observations were seen when a real solvent (cyclohexane) was used as the volatile component instead of air.

The experimental foaming results indicate that vacuum (at a level of supersaturation conditions) is not enough to boil a viscous solution even in the presence of bubbles (nucleation sites), but shear also has to be applied. Furthermore, there is perhaps a critical shear rate or a critical shear stress at which boiling appears. These observations are also in agreement with the Lee and Biesenberger theory for heterogeneous bubble nucleation in polymer melts, as discussed in the first part of this article.

Bubble growth experiments

Figure 4 shows a bubble growth experiment in which the absolute pressure over the liquid was 94 mmHg. At a low shear rate of 3.1 1/s we decided to follow a single bubble (see Figure 4a). The same bubble, but minutes later, is shown in Figure 4b and 4c. Bubble growth cannot be explained by the decrease in the hydrostatic pressure as it rises, since this effect is negligible at this relatively high pressure. Therefore, the bubble growth is a consequence of mass transfer.

Then, a high shear rate of 14 1/s was applied. Our bubble, shown in Figure 4d, is of course more elongated now, since the capillary number increased. The growth (and rise) phenomenon of the bubble is seen in Figure 4e and 4f. Note that in this experiment at relatively high pressure, a weak foaming line starts to appear close to the bottom of the cylinders (see Figure 4f).

Another experiment is shown in Figure 5, where the absolute pressure above the liquid surface was only 9 mmHg. Note that this time the growth rate of the bubble is much faster and that at the higher shear rate, boiling occurred at the bottom and at the top. From the (bubble growth) experimental results, we found that the rate of change of the half-length of

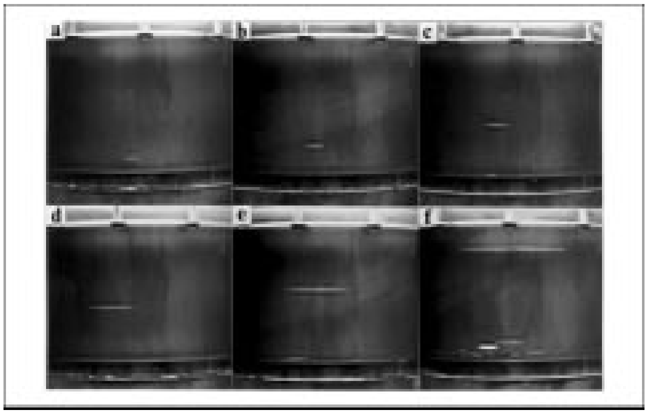

Figure 4. Bubble growth at 94 mm Hg.

(a)–(c): $G = 3.1$ 1/s; (d)–(f): G = 14 1/s. (a) $t = 0$ s, L = 0.60 cm; (b) $t = 672$ s, $L = 0.95$ cm; (c) $t = 1367$ s, $L = 1.2$ cm. (d) $t = 0$ s, $L = 2.2$ cm; (e) $t = 565$ s, $L = 3.05$ cm; (f) $t = 1304$ s, $L = 4.8$ cm.

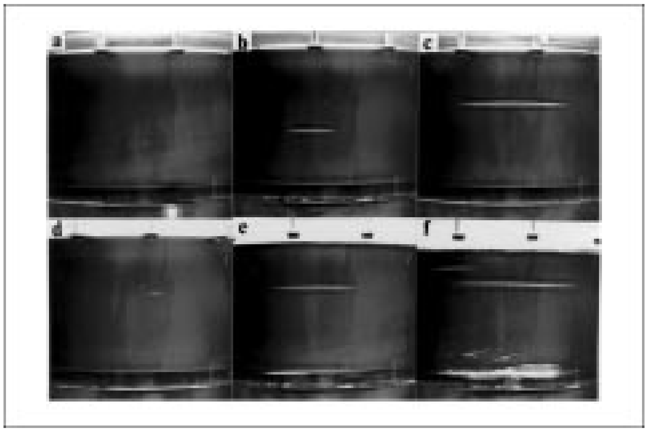

Figure 5. Bubble growth at 9 mm Hg.

(a)–(c): $G = 3.2$ 1/s; (d)–(f): $G = 14$ 1/s. (a) $t = 0$ s, $L = 0.235$ cm; (b) $t = 241$ s, $L = 2.25$ cm; (c) $t = 513$ s, $L = 5.15$ cm. (d) $t = 0$ s, $L = 0.80$ cm; (e) $t = 99$ s, $L = 3.95$ cm; (f) $t = 178$ s, $L = 6.0$ cm.

the bubble is almost constant, that is, it almost does not depend on time or on the half-length of the bubble.

Figure 6 shows a log–log plot of the constant rate of change of the half-length of an air bubble as a function of the shear rate at different ambient pressures. Note that because of the very difficult experimental procedure, experiments at intermediate shear rates have not been done. Therefore, it was decided to concentrate on more experiments with a fewer number of points, at different operating pressures, rather than trying to get more points at the same pressure.

Returning now to Figure 6, it can be seen that the growth rate of the bubble's half-length increases as the shear rate increases and as the pressure decreases. Those two observations are in agreement with the simple model. The slope of the lines should approximately represent the power over the shear rate (G), which according to Eq. 9 should be 3/4. From Figure 6 the average slope is $0.73 \pm 30\%$, a value that is extremely close to the theoretical value of 3/4 suggested in the simple model. The relatively large spread of the experimental results cannot be explained by the error in measuring the bubble's growth rate (estimated to be below 5%). The spread is instead a result of the theoretical and experimental complex problem of bubble growth in a simple shear flow, as discussed in this work.

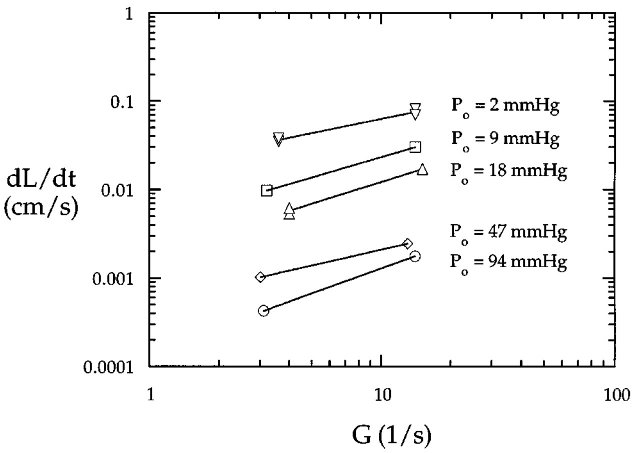

Figure 6. Rate of change of the half-length of the bubble as a function of the shear rate at different ambient pressures.

Conclusions

The results presented here for the nucleation and growth of a bubble in a Newtonian liquid in a simple shear and creeping flow indicate that supersaturation conditions are not sufficient to boil a viscous solution, even when nucleation sites are present, as shear (above a certain level) also has to be applied. Furthermore, the growth rate of a slender bubble increases as the shear rate increases and as the pressure decreases. These results probably can explain higher mass-transfer rates obtained in rotary machines such as vented extruders, at higher rotational speeds.

Acknowledgment

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