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## Marangoni Effects on the Bubble Dynamics in a Pressure Driven Flow

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### ABSTRACT

The motion of air bubbles and water drops in a Hele-Shaw cell filled with a silicone oil has been studied experimentally and theoretically. By adding a predetermined amount of a surfactant to the water drops we attempted to investigate the surfactant influence systematically. While the motion of air bubbles was in reasonable agreement with the predictions of Taylor and Saffman [Q. J. Mech. Appl. Math. 12, 265 (1959)], water drops behaved quite differently in that the translational velocities were smaller by an order of magnitude and their shapes were very unusual as observed previously by Kopf-Sill and Homsy [Phys. Fluids 31, 18 (1988)]. Assuming that the surrounding fluid wets the solid wall and the bubble (or the drop) surface is rigid due to the surfactant influence, we have estimated the translational velocity of an elliptic bubble. The calculated velocities were in good agreement with the observations indicating that the surfactant influence could retard the bubble motion significantly. The present study also indicates that the unusual bubble shapes are also due to the surfactant influence.

### INTRODUCTION

Various physical systems involving the motion of bubbles or drops often show many unexpected flow phenomena which cannot be explained by existing theories. The unexpected flow characteristics of bubbly flows may include unusually high pressure drops, low bubble velocities, and perplexing bubble shapes. Furthermore, these flow characteristics are usually bubble-size dependent. While many of the unexpected flow phenomena may be attributed to the influence of surface active substances present in the system, details of such flows are yet to be fully understood.

When bubbles or drops are driven by a surrounding immiscible fluid, the flow may involve two characteristic velocities; the bubble velocity and the average velocity of the surrounding fluid. Although the bubble is driven by the surrounding fluid, the two characteristic velocities are not necessarily the same depending on the flow situations. If surface active substances are present, the flow becomes even more complicated since they affect the relative magnitude of the two characteristic velocities, resulting in various flow regimes. Although these complex flow situations may be encountered whether the gravity effect is present or not, the fundamental issue is in understanding the balance between the surface elasticity induced by the non-uniform surface tension and the viscous force under various flow conditions. Thus, in studying such flows it is desirable to minimize the gravity influence which introduces unnecessary complication. In ground-based experiments, the gravity effect may become negligible in a flow where the

characteristic length scale is very small. Examples may include the motion of bubbles in a capillary tube or in a Hele-Shaw cell. The motion of bubbles in a Hele-Shaw cell is especially rich in exhibiting various perplexing flow characteristics which are apparently caused by the surfactant influence.

The motion of a finite bubble moving in a Hele-Shaw cell which is filled with a viscous liquid was first analyzed by Taylor and Saffman (1959). Neglecting the surface tension and the wetting film between the bubble and the plates (Fig. 1), they predicted that a small elliptic bubble with its longer axis in the flow direction moves with a velocity  $U$  greater than  $2V$ . Here  $V$  is the average velocity of the surrounding fluid which drives the bubble (Fig. 1). When  $U < 2V$ , on the other hand, the shorter axis of the elliptic bubble is in the flow direction. If the bubble is circular,  $U=2V$ . In any case their solution indicates that the bubble velocity is always greater than the average velocity of the surrounding fluid (i.e.,  $U > V$ ).

Maxworthy (1986) conducted an experiment with bubbles that were driven by buoyancy rather than by a pressure gradient. He investigated air bubbles moving in an inclined Hele-Shaw cell which was filled with silicone oil. The bubbles in his experiment were ellipses elongated in the flow direction. While large bubbles moved somewhat faster than the Taylor-Saffman prediction, small bubbles moved slower than expected. Kopf-Sill and Homsy (1988) conducted similar experiments, but theirs was a pressure driven flow in which air bubbles in a horizontal Hele-Shaw cell were driven by a glycerin-water mixture. Unlike the experiments of Maxworthy, they observed a variety of unusual bubble shapes such as ovoids, long- and short-tailed bubbles. In addition, the bubble velocities were much smaller than expected by an order of magnitude. Considering the fact that aqueous systems are very prone to contamination by surface active substances, it may be possible that the perplexing observations by Kopf-Sill and Homsy may be due to the influence of surface-active contaminants.

In this paper, the results of an experimental study are presented in which water drops containing sodium dodecyl sulfate at a predetermined concentration were driven through a Hele-Shaw cell filled with a silicone oil. A theoretical calculation is also presented for the bubble velocity which accounts for the influence of surfactants. Both experimental and theoretical results show reasonable agreement with those of Kopf-Sill and Homsy indicating that surface active contaminants can be responsible for the observed retardation and for the perplexing bubble shapes.

## EXPERIMENTS

The Hele-Shaw cell consisted of two 1/2-in thick Pyrex glass plates separated by a rubber gasket of 0.9 mm or 1.8 mm in thickness. The effective cell dimension was 17.8 cm by 86.4 cm. A silicone oil with the measured viscosity and surface tension of 97 cp and 21 dyn/cm was used as the driving fluid. The cell had an injection port at one end of the top plate so that an air bubble or a water drop could be introduced into the cell using a syringe. The planform diameter of bubbles and drops was controlled to be at about 1.3 cm and 2.1 cm throughout the experimental

study. Water drops containing a surfactant at various concentrations were prepared by dissolving sodium dodecyl sulfate (SDS) in distilled water at a concentration of 5%, 10% and 20% of the critical micelle concentration (CMC), respectively. The CMC of SDS in water at 25°C is 8.2 mmol/l which is equivalent to 0.236% by weight. The interfacial tension at room temperature was 34.3, 32.1 and 25.9 dyn/cm at the prescribed concentrations of SDS.

Once an air bubble (or a water drop) of predetermined size was positioned at one end of the cell, the silicone oil was driven by a peristaltic pump at various flow rates to induce the bubble motion. The translational velocity and the shape of the bubble (or drop) were then recorded for each value of the oil flow rate. The measured velocities are plotted as a function of capillary number  $Ca$  in Fig. 2.  $Ca$  is defined as  $\mu U/\sigma$  where  $\mu$  is the viscosity of the surrounding fluid,  $U$  is the bubble velocity and  $\sigma$  the equilibrium value of the interfacial tension. Although some discrepancies existed, the air bubbles were moving with a velocity close to the prediction of Taylor and Saffman and their shapes were near circular with slight elongation in the flow direction. Water drops, on the other hand, were moving much slower than predicted, and they were elongated in the transverse direction unlike the air bubbles. At the prescribed concentrations of SDS, the data sets were indistinguishable from one another. Although not indicated in the figure, some drops showed a transition to a short-tailed shape at a higher capillary number in accordance with the observations of Kopf-Sill and Homsy. These differences between air bubbles and water drops are apparently due to the surfactant influence as supported by the theoretical calculation given in the following section. More details of the experimental study can be found in Park et al. (1994).

## ESTIMATION OF BUBBLE VELOCITY

In the absence of surface tension effect, the analysis of Taylor and Saffman predicts an elliptic shape when the bubble size is much smaller than the width of the cell. If surface active substances are present, the bubbles are not necessarily elliptical and may take on various interesting shapes depending on the flow condition. Nevertheless, we assume an elliptic plan form since the shape distortion is small at a low capillary number as observed experimentally. While the bubble shape should be determined as a part of solution, the current analysis is an approximation in which the bubble shape is assumed *a priori* in order to obtain an analytic progress.

As indicated in Figure 1, the two principal axes of the elliptic bubble are assumed to be aligned with the flow and the transverse directions with  $2a$  and  $2b$  denoting the bubble dimensions in the two principal axes. The surrounding fluid wets the solid surface thus forming a thin liquid film between the plates and the bubble. In the presence of surface active substances, the Marangoni effect resulting from the surface tension gradient may complicate the flow field near the bubble. When the bubble is small, however, it may be simplified since the entire bubble surface may become rigid (Davis & Acrivos, 1966; Park, 1992). Here we consider the case of a small bubble in which the entire bubble surface is assumed to be rigid.

The flow field slightly away from the bubble is known to be parabolic in the  $xz$ -plane (i.e., across the gap). Furthermore, it can be described as a potential flow in the  $xy$ -plane. Thus,

$$\mathbf{v} = 1.5 \mathbf{v}_{avg} \left( 1 - \frac{4z^2}{h^2} \right) \quad (1)$$

where  $\mathbf{v}_{avg}$  is the gap (or depth)-averaged velocity field described by the potential flow in  $xy$ -plane. In an elliptic cylindrical coordinate system  $(\zeta, \eta)$ , the complex potential  $\Omega$  for the flow past an elliptic cylinder in a bubble fixed frame of reference is given by

$$\Omega = \Phi + i\Psi = -b(V-U) \frac{(k+1)}{2} \sqrt{k^2-1} \left[ \frac{e^\gamma}{k+1} - \frac{e^{-\gamma}}{k-1} \right] \quad (2)$$

where  $\gamma = \xi + i\eta$  and  $k$  the shape factor defined as  $k = a/b$ .  $V$  and  $U$  are the magnitude of the average velocity of the surrounding fluid and the bubble, respectively. From the gradient of the velocity potential  $\Phi$  or the stream function  $\Psi$ , the velocity components in  $\xi$  and  $\eta$  directions (hence  $\mathbf{v}_{avg}$ ) can be obtained.

In the thin film region between the plates and the bubble, the flow field is represented by a Couette flow if the bubble surface is rigid. The previous analyses on the bubble motion in a capillary tube or in a Hele-Shaw cell provide an expression for the film thickness which is proportional to  $Ca^{2/3}$  (Park, 1992). Thus, the flow field in the thin film region is known. Eqns (1), (2) and the Couette flow in the film region represent the three-dimensional velocity field around the translating bubble excluding the small region of  $O(h)$  in the immediate vicinity the bubble rim. Using this velocity field, an analytic description of the translational velocity of the elliptic bubble can be obtained from an integral form of the  $x$ -directional momentum balance as follows:

$$F_D = \frac{12\mu}{h} \frac{\pi c^2}{4} \left\{ V \frac{k+1}{k-1} - \left[ (V-U) + U \frac{k-1}{k+1} \right] + (V-U) \frac{2}{k-1} \right\} - \frac{16\mu U(abI)}{2.122hCa^{2/3}} \quad (3)$$

Here  $c = \sqrt{|a^2 - b^2|}$  and  $I$  is a constant given as a definite integral which accounts for the film thickness variation in the transverse direction (i.e.,  $y$ -direction) for a given value of  $k$ .  $I$  varies from 0.79 to 1.29 for  $k$  changing from 0.1 to 3.0. For a circular bubble (i.e.,  $k = 1$ ),  $I = 0.91$ .  $F_D$  is the drag force acting on the bubble which is zero for a freely suspended drop. Consequently, the following expression for the bubble velocity can be obtained:

$$U = \beta(k+1)V \quad \text{where} \quad \beta = \frac{Ca^{2/3}}{Ca^{2/3} + 0.2kI} \quad (4)$$

Here  $\beta$  is the retardation factor introduced by the surfactant influence which is  $O(Ca^{2/3})$ . This order of magnitude decrease in the bubble velocity is due to the large drag in the thin film region where the bubble surface is rigid. Since this drag is proportional to the film thickness, the bubble velocity is also proportional to  $Ca^{2/3}$ . In the absence of Marangoni effect, the bubble surface in the thin film region is stress free, and the expression for the bubble velocity is equivalent to setting  $\beta = 1$ . Thus the result of Taylor and Saffman for elliptical bubbles is recovered.

In Figure 2, the calculated  $U/V$  using Eqn (4) has been overlaid for various value of  $k$  to compare with experimental results. It appears that the measured velocities are in reasonable agreement with the current predictions for ellipses which are elongated in the transverse direction (i.e.,  $k < 1$ ). While the drops in the experiment were not exactly elliptical, they were nearly elliptical and elongated in the transverse direction. Experiments indicate that the drops are elongated sideways at a low capillary number and their aspect ratio decreases with increasing capillary number. It is interesting to note that the data points show better agreement with the theoretical curve with smaller  $k$  at a lower  $Ca$ . With increasing  $Ca$ , the agreement becomes progressively better with the curves for larger  $k$  in accordance with the experimental observations.

## CONCLUSIONS

The present study indicates that surface active contaminants can influence the bubble dynamics significantly, and that most of the perplexing observations by Kopf-Sill and Homsy are probably due to the influence of surface active substance which may be present in the system as contaminants. As Kopf-Sill and Homsy anticipated, the significant retardation of bubble velocity and remarkable bubble shapes, in fact, result from the three-dimensional nature of the flow which cannot be explained by the two-dimensional Hele-Shaw equations.

## ACKNOWLEDGMENTS

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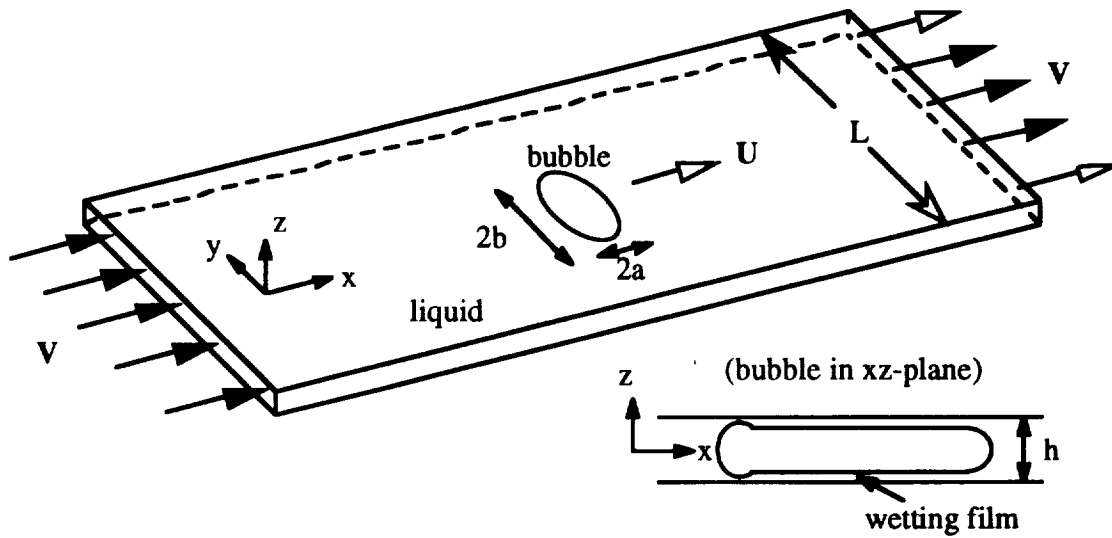


Figure 1 Schematic of a bubble moving in a Hele-Shaw cell

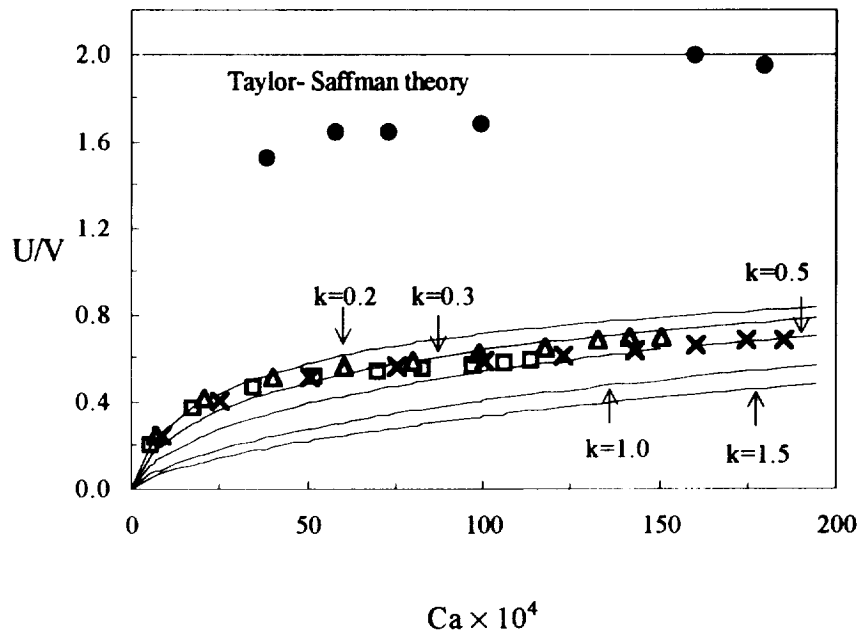


Figure 2 Comparison of experimental results and the theoretical estimate given by Eqn. (4) (●, air bubbles; Open symbols are for water drops at the SDS concentration of 5% (□), 10% (Δ), and 20% (×) of CMC.)