SURFACTANT EFFECT ON RISING BUBBLES – A THIRD ORDER THEORY

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We study the surfactant effects on the motion of an air-bubble rising in a vertical capillary tube of small radius R, filled with a viscous fluid and sealed at one end. The thickness b of the thin film behind the bubble is small compared with R. We give a theoretical estimate of b, by using an expansion of order $O(b/R)^3$ of some functions appearing in the mathematical model. An upper bound of the rising velocity U is given in terms of b and other physical data of the problem. The obtained estimates are proving the thinning and the delay effect due to the surfactant presence on the bubble interface: b and U are smaller, compared with the "clean" case. Our results are consistent with previous experimental and numerical data. In the case of horizontal (infinite) capillary tubes we have the opposite effect of surfactant, proved theoretically by Daripa & Paşa (2010).

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1. INTRODUCTION

Gas-bubbles flow through viscous fluids was studied in many papers over the years, related with fundamental and technological problems. Davies and Taylor (1950) studied this problem, related with the submarine technology. Saffman and Taylor (1958) obtained important results in this problem, related with oil recovery technology and fingering phenomena in displacement processes. The fingering problem in Hele-Shaw displacement was studied also by Park and Homsy (1984). Experimentally studies on the long air-bubbles in vertical cylindrical tubes have been carried on by White and Beardmore (1962) and Zukoski (1966). Potential analysis was used to study two-dimensional bubbles in tubes, by Couet et al. (1985) and Daripa (2000). Schwartz et al. (1986) studied the displacement of a viscous fluid by a gas-bubble in capillary tube. Fabre and Line (1992) given a review of long bubble propagation in capillary tubes. The case of angular capillary tubes was considered by Bico & Quere (2002). Bubbles flow in non-circular cross-section tubes has been studied by Liao & Zhao 2003 and Clanet et al. (2004). In all the above papers, the surface tension γ on the bubble interface was considered *constant*.

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We consider here a long air-bubble rising with a velocity U in a vertical tube of small radius R. In the case of a constant surface tension γ on the bubble interface, a seminal paper was given by Bretherton (1961). Reinelt (1987) reinforced the results of Bretherton. The tube is filled with a liquid of viscosity μ and is sealed at one end. In both papers was pointed out that the shape of the cap and the base of the bubble are independent of the size of the bubble. The bubble can be considered almost cylindrical, with radius βR. The thickness of the layer behind the bubble, between the wall of the tube and the bubble interface, is $b = R - \beta R = R(1-\beta)$. The front meniscus of the air-bubble is displacing a quantity of fluid. The tube being sealed, this quantity of liquid must flow down, under the effect of gravity, exactly through the thin layer between the bubble interface and the tube wall.

We study here the above problem in the case of a *variable* surface tension γ, due to the small traces of surfactant on the bubble interface.

The important forces acting in this problem are: surface tension, viscous force, gravity and inertia. Associated with these forces we have the Reynolds number Re, the capillary number Ca and the Bond number B given by

$$Re = \rho U R/\mu, \quad Ca = \mu U/\gamma, \quad B = \rho g R^2/\gamma,$$

where ρ is the density of the fluid neglecting the density of the air and g is the gravitational acceleration. At a constant rising velocity U, the experiment of Bretherton (1961) suggests that Reynolds number is negligible, then only parameters Ca and B appear in the problem. Bretherton (1961) used matched asymptotic expansion in terms of small Ca and obtained the expansion

$$B - 0.842 \sim 1.25 Ca^{\frac{2}{3}} + 2.24 Ca^{\frac{1}{3}}, \tag{1}$$

therefore the bubble is not rising if $B < 0.842$.

A very simple estimate of the values of B which allows the rising of a spherical bubble can be obtained in terms of gravitational and surface tension. The bubble is pushed up by the force $(4\pi R^3/3)\rho g$. On the other hand, the surface tension γ is acting on the circumference $2\pi R$, then the total "opposite" force is $2\pi R\gamma$. The rising conditions is

$$\frac{4\pi R^3}{3}\rho g > 2\pi R\gamma \Rightarrow B > 1.5, \tag{2}$$

and we see that is independent of the capillary number Ca. In the case of a long bubble, considered here, we don't know the total volume of the bubble, then the above idea is not useful. However, the formula (2) is an "empirical" justification for the existence of a rising condition in terms of the Bond number B.

White and Breadmore (1962), using some experimental data (cited also by Clanet et al. (2004)), obtained the relation

$$U = C \cdot \rho g R^2/\mu, \tag{3}$$

that means $B = C^{-1}(Ca)$, whit $C \approx 0.038$. The value of value of the constant C monotonically decreases with decreasing Bond number B.

Even in the paper of Bretherton (1961) was pointed out that some theoretical results are not in good agreement with the experimental data – the experimental thickness of the thin film was smaller, compared with the theoretical values. The formula (1) was not verified qualitatively by Bretherton in the case $Ca < 10^{-5}$. He considered the quantitative discrepancies as due to the traces of surfactant on the bubble surface, which are giving a thinning effect on the thin layer behind the bubble. The basic models for the surfactant effects on the fluid flow can be found in the book of Levich (1962). As we pointed above, the main result of our paper is a theoretical explanation of this disagreement. We give a third order precision theory, neglecting the terms $O(b^4)$, and obtain the thinning and the delay effect produced by the surfactant presence on the bubble interface. The thinning effect of surfactant in the case of rising bubbles was confirmed by the numerical and experimental results of Amatroushi & Borhan (2004). The surface tension is considered variable, in terms of the surfactant concentration on the bubble surface. We consider here that the surfactant is not soluble in the bulk (the viscous fluid in the tube).

We emphasize that in the case of horizontal tubes, the opposite effect, due also to the surfactant presence on the bubble interfaces, was supposed by Bretherton. The case of bubble flow in in horizontal capillary tubes is totally different. There it has been proved that the interfacial surfactant thickens the thin film in comparison to the clean case. This case is quite similar with the Landau–Levich coating problem: a plate is moving out from a bath of viscous liquid and a thin layer is adhering on it. The traces surfactant on the bath surface are giving the same thickening of the adhering film. In both problems we mention the theoretical contributions of Daripa & Paşa (2009, 2010) and numerical and experimental contributions of Park (1990) and Ratulowski & Chang (1989, 1991).

The vertical case which we study here is different. As we pointed above, the liquid displaced by the front meniscus of the rising bubble must flow back down exactly through the thin film between the bubble surface and the tube walls, under the effect of gravity. On the contrary, in the horizontal tubes, the thin film is almost at rest and *the gravity effect is neglected.*

The paper is laid out as follows. In Section 2 we give an approximate value of b, U in the clean case. The surfactant effect is described in Section 3 and the thinning and the delay effects are proved. Finally, we conclude in Section 4.

2. THE APPROXIMATE VALUES OF b, U IN THE "CLEAN" CASE

In Figure 1 is given a 2D axisymmetric gas-bubble in a capillary tube. We use first a Cartesian frame: the x-axis is the right wall of the tube and is pointed upward. The y-axis is orthogonal to the right wall and pointed to the tube center. The gravity is parallel with the x-axis and pointed downward. b denotes the (constant) thickness of thin liquid layer between the bubble surface and the tube walls, U is the rising velocity and R is the tube radius. The bubble surface is "divided" in three regions: the region AB of the front meniscus, the intermediate region BC and the flat region CD, where the film thickness is constant. The free surface of the bubble in the transition region BC is denoted by $h(x)$. In this section, we consider the case of a constant γ, i.e., the clean case.

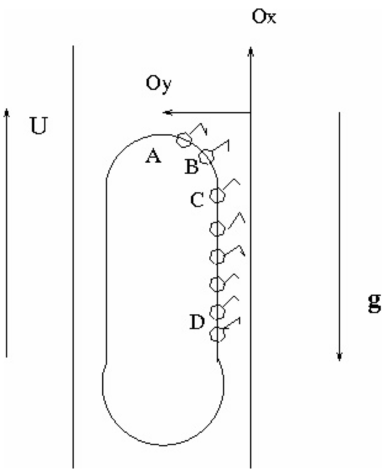

Fig. 1. Section of a bubble in a vertical tube. Cartesian co-ordinates.

In the transition zone BC, we use the lubrication approximation

$$u_{yy} = \frac{1}{\mu}(p_x + \rho g), \quad p_y = 0. \tag{4}$$

Both above equations must be solved subject to the following boundary conditions for the "clean" case (see also Daripa and Paşa, 2010)

$$u(y = 0) = 0, \quad u_y(y = h(x)) = 0, \quad \text{and} \quad p(y = h(x)) = -\gamma h_{xx}. \tag{5}$$

The solution of the problem (4)–(5) is given by

$$u = \frac{1}{\mu}(p_x + \rho g)(y^2/2 - yh). \tag{6}$$

The flux $Q(x)$ in the thin layer behind the bubble in the point $x \in BC$ is given by

$$Q(x) = \frac{1}{\mu}(p_x + \rho g)(-h^3/3). \tag{7}$$

The matching procedure, used by Bretherton (1961), means that the solution (6) can approximate the flow also near the point C for the flat region. Therefore we use the above expression of the flux near the point C where the curvature h_{xx} of the free surface of the layer becomes zero and $h \approx b$. The thickness of the bubble is constant in the flat region. Since $Q(C) = Q(-\infty)$ due to incompressibility, we obtain

$$Q(-\infty) = -\frac{\rho g b^3}{3\mu}. \tag{8}$$

On the other hand, in the front of the bubble we have a flux of fluid with speed U through the area of radius $(R - b)$. The tube is sealed, therefore the displaced fluid must flow down, through the thin layer between the bubble surface and the tube walls. Then we obtain the following second expression for the flux per unit (circumferential) length

$$Q(\infty) = -U[\pi(R-b)^2]/(2\pi R) \approx -\beta^2 \frac{UR}{2}, \tag{9}$$

where

$$\beta = 1 - b/R = 1 - \delta. \tag{10}$$

Since $Q(-\infty) = Q(\infty)$ we get the following expression of U obtained by Bretherton (1961)

$$U = \left(\frac{\rho g R^2}{\mu}\right) \frac{2(1-\beta)^3}{3\beta^2} = \left(\frac{\rho g R^2}{\mu}\right) \frac{2\delta^3}{3(1-\delta)^2}. \tag{11}$$

We consider now the cylindrical co-ordinates used by Reinelt (1987), for describing the axisymmetric flow in the transition region. The Ox axis is now the upward symmetry axis of the tube (which is different from the convention used in the Cartesian frame before) and r is the radial distance from the tube center. The gravity force is downward. The flow equations in the transition region BC are

$$\frac{1}{r}\frac{d}{dr}(ru_r) = \frac{1}{\mu}(p_x + \rho g), \quad p_r = 0, \tag{12}$$

whit boundary conditions

$$u(r = R) = 0 \quad \text{and} \quad u_r(r = R - b) = 0. \tag{13}$$

The solution of the the problem (12)–(13) in the flat region CD, where $p_x = 0$, is given by

$$u(-\infty, r) = -\frac{\rho g}{4\mu}\{R^2 - r^2 + 2(R-b)^2 \ln(r/R)\}. \tag{14}$$

Therefore, the flux far downstream (far from the front meniscus, i.e., through the flat region CD) is then

$$Q(-\infty) = \int_0^{2\pi} \int_{R-b}^{R} u(-\infty, r) r \,\mathrm{d}r \,\mathrm{d}\theta = -\frac{\pi \rho g}{8\mu} R^4 \, E(\beta), \tag{15}$$

where

$$E(\beta) = 1 - 4\beta^2 + 3\beta^4 - 4\beta^4 \ln(\beta). \tag{16}$$

As in the above formula (9), the liquid displaced by the front meniscus of the bubble (with rising velocity U), must flow down through the thin film, then we get the second expression of the flux

$$Q(\infty) = -2\pi \int_{R-b}^{R} U r \,\mathrm{d}r = -U\pi(R-b)^2. \tag{17}$$

Equating (15) and (17) and simplifying, we obtain the formula of Reinelt (1987)

$$U = \frac{\rho \, g \, R^2}{\mu} \left(\frac{E(\beta)}{8\beta^2} \right) = \frac{\rho \, g \, R^2}{\mu} \left(\frac{E(1-\delta)}{8(1-\delta)^2} \right). \tag{18}$$

Remark 1. Consider the segment $\beta \in (0, 1)$, then $E(\beta)$ given by (16) is positive. Indeed, we have

$$E'(\beta) = (-8\beta)D(\beta), \quad D(\beta) = 1 - \beta^2 + 2\beta^2 \ln(\beta), \quad D'(\beta) = (4\beta)\ln(\beta),$$
$$D'(\beta) < 0, \; D(0) = 1, \; D(1) = 0 \Rightarrow D(\beta) > 0.$$

It follows $E'(\beta) < 0$, then $E(\beta)$ is strictly decreasing function. We have $E(0) = 1$, $E(1) = 0$, therefore, $E(\beta) > 0$ in the segment $\beta \in (0, 1)$. $\square$

Both formulas (18) and (11) are giving *approximate relations* between the rising velocity U and the undimensional film thickness $\delta = b/R$, because we used the lubrication approximation. As we pointed above, we expect a small value for δ. Therefore, in a small interval near the origin, we expect that (18) and (11) are giving two *exact relations* between U and δ. From here we obtain an approximate value of b, as follows.

In the formula (16) of the function $E(\beta)$ is appearing $\ln(\beta)$. As we expect to have $\delta < 1$, we can use an expansion of this last function in terms of δ. Recall (10). We use the third order expansion, because in (11) only δ^3 appears. We have

$$\ln(\beta) = \ln(1-\delta) \approx -\delta + \delta^2/2 - \delta^3/3, \tag{19}$$

$$3E(1-\delta) \approx 3\left[1 - 4(1-\delta)^2 + (1-\delta)^4\{3 - 4(-\delta + \delta^2/2 - \delta^3/3)\}\right] = \\ = -12\delta^2 + 64\delta^3 + O(\delta^4). \tag{20}$$

Consider the following polynom $P(\delta)$:

$$P(\delta) = -12\delta^2 + 64\delta^3 - 16\delta^3 = 12\delta^2(4\delta - 1), \tag{21}$$

therefore, $3E(1-\delta) - 16\delta^3 = P(\delta) + O(\delta^4)$. By equating (18) and (11) in the third approximation (that means neglecting the terms $O(\delta^4)$) we get $P(\delta) = 0$, therefore

$$\delta \approx 0.25, \quad \beta \approx 0.75, \quad b \approx R/4. \tag{22}$$

The above theoretical estimate was obtained by using the expansion (19). It is not an exact value of b, but we use it to prove the thinning effect of surfactant. The expansion of $\ln(1-x)$ is accurate only for small value of x. We give here some numerical values of error function $ERR(x) = \ln(1-x) - (-x + x^2/2 - x^3/3)$:

(23)

$$ERR(0.25) = -0.063723, \ ERR(0.2) = -0.040476, \ ERR(0.1) = -0.010027.$$

Therefore, in our case the relative error is $0.05/0.25 \approx 20\%$.

We use the result (22), the formula (11) and obtain the approximate value of the rising velocity U

$$U \approx 0.0184\left(\frac{\rho g R^2}{\mu}\right). \tag{24}$$

We can see that the value 0.0184 is less than the experimental value $C = 0.038$ (see White & Beardmore (1962)). Thus our approximation is underestimating the rising velocity in comparison with the experimental value.

3. THE THINNING EFFECT OF SURFACTANT

In the presence of interfacial surfactant with no surfactant in the bulk, the surfactant concentration on the interface will be not constant. Due to the motion of the bubble, surfactant along the interface will be swept downstream along the interface. This will cause surfactant concentration to increase and hence surface tension γ to decrease away from the front end of the bubble along its interface. Since the Ox axis is upward (see Figure 1), it follows that $\gamma_x > 0$ along the interface. The thickness of the layer between the bubble and the wall of the tube will be denoted here by b_S and the rising velocity by U_S (subscript S stands for surfactant case). We use the notations

$$\beta_S = 1 - b_S, \quad \delta_S = b_S/R.$$

As before, the equations in the transition region BC are same as (4) but the boundary conditions are now given by the relations (see Daripa & Paşa (2010), Ratulowski & Chang (1990), Park (1991))

$$u(y=0)=0, \quad u_y(y=h(x))=\frac{\gamma_x}{\mu} \quad \text{and} \quad p(y=h(x))=-\gamma h_{xx}. \tag{25}$$

The solution of the system (4)–(5) is given by

$$u=\frac{1}{\mu}(p_x+\rho g)(y^2/2-yh)+\frac{\gamma_x}{\mu}y. \tag{26}$$

From this, the flux $Q(x)=\int_0^{h(x)}u(y)\mathrm{d}y$ through the film at an arbitrary point x in the transition region BC is obtained as

$$Q(x)=\frac{1}{\mu}(p_x+\rho g)(-h^3/3)+\frac{\gamma_x}{\mu}\frac{h^2}{2}. \tag{27}$$

The solution (26) and the flux (27) are valid in the transition region and hence they hold approximately in the overlap region with the flat region. Consider a point C^- situated below and near the point C. The curvature $h_{xx}(C^-)$ of the free surface at the point C^- is approximately zero and $h(C^-)=b_S$ (since thickness of the film is constant in the flat region). Moreover, in general γ_x is not zero in the thin film region due to the tendency of surfactant to get advected downstream. Therefore, $\gamma_x(C^-)\neq 0$ and flux through the point C^- behind the front meniscus is given by

$$Q(C^-)=-\frac{\rho g b_S^3}{3\mu}+\frac{\gamma_x(C^-)}{\mu}\frac{b_S^2}{2}. \tag{28}$$

The front meniscus of the bubble is displacing a quantity of fluid with the rising velocity U_S on the area of radius $(R-b_S)$. Then we have the second expression of the flux through the thin layer behind the bubble, given by the formula (9) with U_S instead of U. Equating (28) and (9) with U_S instead of U (that means mass conservation), we get

$$U_S=\left(\frac{\rho g R^2}{\mu}\right)\frac{2(1-\beta_S)^3}{3\beta_S^2}-\frac{\gamma_x(C^-)R}{\mu}\cdot\frac{(1-\beta_S)^2}{\beta_S^2} \tag{29}$$

Remark 2. The above formula gives us a criterion for positive rising velocity

$$U_S>0 \Leftrightarrow \gamma_x(C^-)<\frac{2R(1-\beta_S)\rho g}{3}. \quad \square \tag{30}$$

Comparing (11) with (29) (recall $\gamma_x(C^-)>0$), we obtain

$$U_S<\left(\frac{1-\beta_S}{1-\beta}\right)^3\left(\frac{\beta}{\beta_S}\right)^2 U. \tag{31}$$

For the axisymmetric flow in the transition region, the governing equations in cylindrical co-ordinates (already introduced previously) are same as (12). The boundary conditions on the flat region of (CD) of the interface are

$$u(r=R)=0, \quad u_r(r=R-b_S)=\frac{\gamma_x}{\mu}, \tag{32}$$

because here the curvature h_{xx} is zero. Then the derivative p_x of the pressure is also zero. As we pointed before, along the interface of the thin film, γ_x is not zero, because the surfactant is swept to the rear meniscus. Recall also that that the bubble is infinite long. From the solution of the problem (12) + (32) with $p_x = 0$ in the thin film region (where the curvature is zero), we get the solution near the point C^- (defined after the relation (27))

$$u(C^-,r) = -\frac{\rho g}{4\mu}\left\{R^2 - r^2 + 2(R-b_S)^2 \ln\left(\frac{r}{R}\right)\right\} + \frac{\gamma_x(C^-)}{\mu}(R-b_S)\ln\left(\frac{r}{R}\right). \tag{33}$$

Therefore, the flux near the point C^-, i.e., through the flat region CD, is given by

$$Q(C^-) = -\frac{\pi\rho g}{8\mu}R^4 E(\beta_S) + \frac{\pi R^3 \gamma_x(C^-)}{2\mu}G(\beta_S), \tag{34}$$

where $\beta_S = (R-b_S)/R$ and

$$G(\beta_S) = -\beta_S + \beta_S^3 - 2\beta_S^3 \ln \beta_S.$$

Far up in front of the bubble, we have a flux of fluid with velocity U_S on the circular area of the radius $(R-b_S)$. This quantity of fluid must flow down through the thin layer of thickness b_S, and the corresponding flux is given by $-U_S\pi(R-b_S)^2$. Equating with the relation (34) we get

$$U_S = \left(\frac{\rho g R^2}{\mu}\right)\frac{E(\beta_S)}{8\beta_S^2} - \frac{\gamma_x(C^-)R}{2\mu}\cdot\frac{G(\beta_S)}{\beta_S^2}. \tag{35}$$

Remark 3. The function $G(\beta_S)$ is positive in $(0,1)$. Indeed, we have

$$G(\beta_S) = \beta_S H(\beta_S), \quad H(\beta_S) = -1 + \beta_S^2 - 2\beta_S^2 \ln \beta_S.$$

Since $dH/d\beta_S = -4\ln(\beta_S) > 0$, $\forall \beta_S \in (0,1)$, we conclude that $H(\beta_S)$ is negative in $(0,1)$, because $H(0) = -1$ and $H(1) = 0$. Therefore, we get

$$G(\beta_S) < 0, \quad \forall \beta \in (0,1). \quad \square \tag{36}$$

We consider that (29) and (35) are giving the same rising velocity U_S, for small b_S, therefore we get

$$\frac{\rho g R^2}{\mu}\left(\frac{E(\beta_S)}{8} - \frac{2(1-\beta_S)^3}{3}\right) = \frac{\gamma_x(C^-)R}{2\mu}\left(G(\beta_S) - 2(1-\beta_S)^2\right). \tag{37}$$

Remark 4. We obtain the thinning effect as follows. The right hand side of the above relation (37) is **negative**, because $\gamma_x > 0$, $G(\beta_S) < 0$ (see (36)) and $(1-\beta_S)^2 > 0$. We recall the polynom $P(\delta)$, given by the relation (21), and from (37) we get

$$3E(1-\delta_S) - 16\delta_S^3 \approx P(\delta_S) = 12\delta_S^2(4\delta_S - 1) < 0 \Rightarrow \delta_S < 1/4 = \delta. \tag{38}$$

This last relation is giving the thinning effect due to the traces of surfactant existing on the bubble surface. □

Remark 5. The delay effect follows from the relation (11) and (29). Indeed, from (29) we get

$$U_S < \left(\frac{\rho g R^2}{\mu}\right) \frac{2(\delta_S)^3}{3(1-\delta_S)^2}, \tag{39}$$

because $\gamma_x > 0$. We also have

$$\delta_S < \delta \Rightarrow \frac{(\delta_S)^3}{(1-\delta_S)^2} < \frac{(\delta)^3}{(1-\delta)^2},$$

therefore the relation (11) is giving $U_S < U$. □

4. CONCLUSIONS

In this paper we study some aspects of the flow of a finite but very long air-bubble rising in a vertical capillary tube which is closed at one end. The fluid displaced by the front meniscus of the bubble must flow down, through the thin layer of thickness b between the bubble surface and the tube wall; the gravity effect is not neglected. We prove analytically that the presence of surfactant on the bubble interface gives a thinning and a delay effect: the thickness of the liquid layer behind the bubble and the rising velocity of the bubble are smaller, compared with the "clean" case (see Remarks 4 and 5). An approximate value of the rising velocity for the clean case is also given which vary widely depending on the radius of the tube as has been also observed in White & Beardmore (1962). Our result is obtained by using the expansion (19), where we neglected the terms $O(\delta^4)$, $\delta = b/R$.

The obtained effects of interfacial surfactant are confirmed by previous experimental and numerical results.

In the case of bubbles moving in horizontal capillary tubes, the fluid in the thin layer behind the bubble is at rest and the gravity is neglected. In this case, the exactly opposite effect of surfactant appears, namely thickening of the thin film (see Bretherton (1961), Park (1991), Ratulowski & Chang (1990), Daripa & Paşa (2010)).

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