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Motion of a gas bubble inside a spherical liquid container with a vertical temperature gradient

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The steady-state motion of a gas bubble inside a non-isothermal, spherical, liquid-filled container is described by taking into account the effects of gravity, the thermally induced gradient of the gas–liquid interfacial tension, and the finite size of the liquid container. The flow fields inside and outside the bubble located at the centre of the container are calculated using a low-Reynolds-number approximation of the fluid equations. The temperature fields are determined by using a low-Prandtl-number approximation of the heat equations. A general expression is obtained for the steady-state migration velocity of the bubble which, under certain conditions, reduces to expressions previously derived by a number of investigators. Finally, an expression for the vertical temperature gradient that will maintain a stationary gas bubble at the centre of the container is formulated.

1. Introduction

The motion of a gas bubble, or in general, a fluid sphere, in an isothermal liquid bath was first investigated, independently, by Rybczynski (1911) and Hadamard (1911, 1912). More recently, Haberman & Sayre (1950) have studied the effect of the container wall on the bubble motion for the particular case where a fluid sphere is initially at a position concentric with a spherical-shell container. Levich (1962) has compared quite thoroughly the terminal velocity of a gas bubble calculated from the Rybczynski–Hadamard formula with that measured from the experiments performed by a number of investigators. The discrepancies between the theoretical predictions and the experimental observations led Frumkin & Levich (1947) to conclude that to correctly determine the drag force on the gas bubble one must take into account the change in the gas–liquid interfacial tension caused by the impurities in the liquid medium. This view has been studied further by Farley & Schechter (1963), and confirmed by Harper, Moore & Pearson (1967). For example, the formula predicting the steady-state migration velocity of a gas bubble in the presence of an interfacial-tension gradient induced by the surfactants is derived in Levich's book (1962) and in a recent paper by Levan & Newman (1976).

In addition to the surfactants, a temperature gradient in the liquid can also induce an interfacial-tension gradient on the gas bubble surface and hence affect the bubble motion. Young, Goldstein & Block (1959), both experimentally and theoretically, have demonstrated this point in their work. Their theoretical treatment is valid, however, only for an unbounded liquid medium. Meyyappan, Wilcox & Subramanian (1981) have derived an expression for the migration velocity of a gas bubble moving toward a planar surface, in a non-uniform temperature field and in the absence of

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gravity. A similar expression for the migration velocity has been obtained for a fluid droplet inside a liquid drop by Shankar, Cole & Subramanian (1981).

The purpose of this work is to study the steady-state motion of a gas bubble inside a spherical liquid container under the influence of gravity, a thermally induced gradient of the gas–liquid interfacial tension, and the finite size of the liquid container (for an equilibrium description of the problem see Mok, Kim & Bernat 1985) – a situation frequently encountered in fabricating high-compression inertial confinement fusion (ICF) targets (see e.g. Kim 1984 and references therein). (Note that a contemporary design of a high-compression ICF target calls for a spherical microshell containing a uniform layer of liquid fusion fuel on the interior surface.) The gas bubble is assumed to be spherical; namely, the energy resulting from the gas–liquid interfacial tension dominates the gravitational energy (for the proof the reader is referred to Mok *et al.* 1985). Another assumption is that the bubble has already attained its terminal velocity when it becomes concentric with the container. The flow velocity fields inside and outside the gas bubble when it is at the centre of the spherical container are obtained by means of a low-Reynolds-number approximation of the relevant fluid equations. The temperature fields are found by evoking a low-Prandtl-number approximation for the heat equations. The expressions for the vertical temperature gradient on the outer surface of the container that will bring about a stationary gas bubble at the centre of the container (namely, a uniform liquid layer on the inner surface of the container) are derived by requiring that the bubble velocity be exactly zero at the centre point.

2. Gas bubble concentric with spherical container

2.1. *Velocity fields*

To determine the flow velocity fields both inside and outside a gas bubble contained in a spherical liquid bath, it is assumed that the fluids under consideration are Newtonian, obey the continuum fluid mechanics, and have constant thermodynamic and transport properties except for the interfacial tension. Since the fluid flows are azimuthally symmetric (there is no reason to believe otherwise), in terms of the Stokes' stream function ψ, the low-Reynolds-number approximation of the fluid equations in the spherical coordinate system is (see e.g. Bird, Stewart & Lightfoot 1960 and Happel & Brenner 1973)

$$E^2(E^2\psi) = 0, \tag{1}$$

where

$$E^2 = \frac{\partial^2}{\partial r^2}+\frac{\sin\theta}{r^2}\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\right),$$

$r = \tilde{r}/R_B$, $\psi = \tilde{\psi}/(R_B\, V_{ref})$, V_{ref} is the reference velocity, and R_B is the radius of the fluid sphere. The tilde denotes that the variables are expressed in real units.

The dimensionless stream function ψ is defined as

$$v_r = -\frac{1}{r^2\sin\theta}\frac{\partial\psi}{\partial\theta}, \tag{2a}$$

$$v_\theta = \frac{1}{r\sin\theta}\frac{\partial\psi}{\partial r}, \tag{2b}$$

where v_r = r-component of the flow, v_θ = θ-component of the flow, $v_r = \tilde{v}_r/V_{ref}$, and $v_\theta = \tilde{v}_\theta/V_{ref}$. Note that with the stream function defined by (2), the continuity

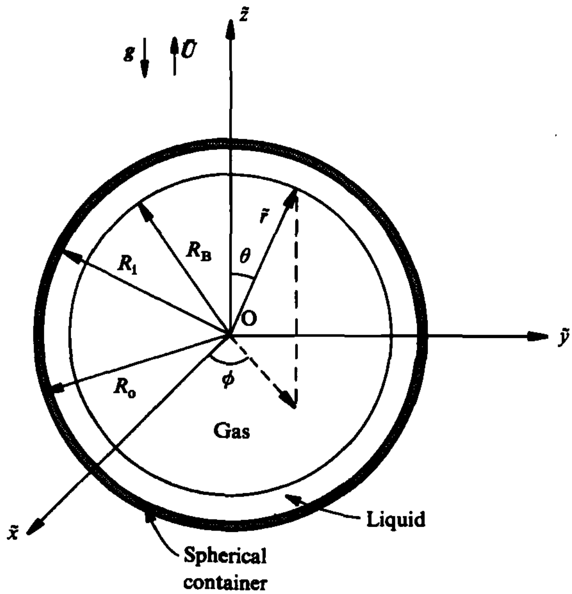

FIGURE 1. Spherical coordinate system used for describing the velocity and temperature fields inside a spherical container. The origin of the coordinate system is at the centre of the gas bubble, which moves upward at a constant velocity $\tilde{U}$.

equation for a fluid with constant mass density, i.e. $\nabla \cdot \boldsymbol{v} = 0$, is automatically satisfied. A spherical coordinate system with its origin at the centre of the gas bubble is shown in figure 1. The bubble (and hence the coordinate system) is assumed to be moving upward at a constant velocity U. The solutions being sought for the velocity fields inside and outside the bubble are of the steady-state type and are valid only at the moment when the bubble is concentric with the outer shell. In general, these kinds of solutions are quasi-static. However, when the bubble is held stationary at the centre of the shell by a vertical temperature gradient, the solutions become truly steady state.

According to Happel & Brenner (1973), the stream functions for the liquid and gas regions which satisfy (1) are, respectively,

$$\psi_\ell(r,\theta) = \sum_{n=2}^{\infty} (A_n r^n + B_n r^{-n+1} + C_n r^{n+2} + D_n r^{-n+3}) \mathscr{I}_n(\cos\theta), \tag{3a}$$

$$\psi_g(r,\theta) = \sum_{n=2}^{\infty} (E_n r^n + F_n r^{-n+1} + G_n r^{n+2} + H_n r^{-n+3}) \mathscr{I}_n(\cos\theta), \tag{3b}$$

where $\mathscr{I}_n(\cos\theta)$ is the nth-order Gegenbauer polynomial.

The eight constants in (3) are determined by the appropriate boundary conditions specified as follows. The non-slip condition at the liquid–solid interface is

$$v_{r,\ell} = -U\cos\theta, \quad \text{at } r = b^{-1}, \tag{4a}$$

$$v_{\theta,\ell} = U\sin\theta, \quad \text{at } r = b^{-1}, \tag{4b}$$

where $U = \tilde{U}/V_{\text{ref}}$, $b = R_B/R_1$, and R_1 is the inner radius of the spherical container.

On the surface of the bubble, i.e. at $r = 1$, the r-component of the fluid flows will vanish because the gas inside the bubble is non-condensable. Specifically,

$$v_{r,\ell} = v_{r,g} = 0. \tag{5}$$

The continuity of the tangential velocity fields requires

$$v_{\theta,\ell} = v_{\theta,\mathrm{g}}. \tag{6}$$

Also, the shear stresses created by the fluid flows and the gas–liquid interfacial-tension gradient should be balanced, i.e.

$$\sigma e_{r\theta,\ell} - e_{r\theta,\mathrm{g}} = -\frac{1}{\mu_{\mathrm{g}} V_{\mathrm{ref}}}\frac{\partial \gamma_{\mathrm{g}\ell}}{\partial \theta}, \tag{7}$$

where

$$e_{r\theta} = \frac{1}{r}\frac{\partial v_r}{\partial \theta} + \frac{\partial v_\theta}{\partial r} - \frac{v_\theta}{r},$$

are the rates of strain in θ-direction, $\sigma = \mu_\ell/\mu_{\mathrm{g}}$, $\mu =$ viscosity of the fluids, and $\gamma_{\mathrm{g}\ell} =$ gas–liquid interfacial tension. The remaining two boundary conditions are the boundedness requirements of the stream function inside the gas bubble.

The eight constants in (3) are determined upon application of the above boundary conditions (4)–(7) and the boundedness of ψ. They are

$$\left.\begin{aligned}
A_n &= L_n b^{2n-3}[(2n-1)-(2n+1)b^2+2b^{2n+1}] \\
&\quad +\delta_{2,n} K_n U[1+\sigma+\tfrac{5}{4}b^3-\tfrac{3}{4}(3-2\sigma)b^5], \\
B_n &= L_n[2-(2n-1)b^{2n-3}+(2n-3)b^{2n-1}] \\
&\quad +\tfrac{1}{2}\delta_{2,n} K_n U[1-(1-\sigma)b^3], \\
C_n &= -L_n b^{2n-1}[(2n-3)-(2n-1)b^2+2b^{2n-1}] \\
&\quad -\tfrac{1}{4}\delta_{2,n} K_n U[(3+2\sigma)b^3-3b^5], \\
D_n &= -L_n[2-(2n+1)b^{2n-1}+(2n-1)b^{2n+1}] \\
&\quad -\tfrac{1}{2}\delta_{2,n} K_n U[3+2\sigma-3(1-\sigma)b^5], \\
E_n &= L_n[2-\tfrac{1}{2}(2n-1)^2 b^{2n-3}+(2n+1)(2n-3)b^{2n-1}-\tfrac{1}{2}(2n-1)^2 b^{2n+1} \\
&\quad +2b^{4n-2}]-\tfrac{1}{4}\delta_{2,n} K_n U\sigma[2-5b^3+3b^5], \\
F_n &= 0, \\
G_n &= -E_n, \\
H_n &= 0,
\end{aligned}\right\} \tag{8}$$

where $\delta_{2,n} = \begin{cases} 1 & \text{for } n = 2, \\ 0 & \text{for } n > 2, \end{cases}$

$$L_n = \frac{1}{4(2n-1)} I_n K_n,$$

$$\frac{1}{K_n} = 1+\sigma-\frac{(2n-1)}{4}(2n-1+2\sigma)b^{2n-3}+\tfrac{1}{2}(2n+1)(2n-3)b^{2n-1} - \frac{2n-1}{4}(2n-1-2\sigma)b^{2n+1}+(1-\sigma)b^{4n-2},$$

$$I_n = -\frac{n(n-1)(2n-1)}{2\mu_{\mathrm{g}} V_{\mathrm{ref}}}\int_0^\pi \frac{\partial \gamma_{\mathrm{g}\ell}}{\partial \theta}\mathscr{I}_n(\cos\theta)\,\mathrm{d}\theta.$$

Following Haberman & Sayre (1950), the drag force on the bubble is given by

$$F_d = -4\pi\mu_\ell V_{\mathrm{ref}} R_{\mathrm{B}} D_n|_{n=2}. \tag{9}$$

Note that only the P_1-mode of the velocity field outside the bubble is responsible for the drag force. The steady-state migration velocity of the bubble is determined by the overall force balance on the bubble, namely the balance among the drag, weight, and buoyancy forces. In dimensionless form, the migration velocity is

$$U = \left[\frac{2}{3\mu_\ell}\frac{R_{\mathrm{B}}^2(\rho_\ell-\rho_{\mathrm{g}})g}{V_{\mathrm{ref}}K_2} - \tfrac{1}{4}\mu_\ell I_2(2-5b^3+3b^5)\right][3+2\sigma-3(1-\sigma)b^5]^{-1}, \tag{10}$$

where g is the gravitational acceleration, and ρ is the mass density.

The function I_2 in (10) represents the effect of the interfacial-tension gradient on the velocity of the bubble. In the present case, the interfacial-tension gradient is created by a non-uniform temperature field so that I_2 can be evaluated once the temperature field is known.

2.2. *Temperature fields*

Assuming that the Prandtl numbers of the fluids are not too high, such that the Péclet number (i.e. the product of Reynolds and Prandtl numbers of the system) is less than unity, one can reduce the heat equation for the fluids to (see e.g. Bird *et al.* 1960)

$$\nabla^2 T = 0, \tag{11}$$

where

$$\nabla^2 = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right)+\frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)$$

for azimuthally symmetric fields and $T = (\hat{T}-T_0)/T_{\mathrm{ref}}$, where T_{ref} is a reference temperature, which is usually chosen as the maximum temperature difference in the system.

This is exactly the Laplacian equation. The heat conduction in the solid wall of the spherical container can also be described by the same equation as long as the properties of the material are constant. The general solutions to (11) in the regions of the wall, liquid and gas are, respectively,

$$T_{\mathrm{w}}(r,\theta) = \sum_{n=1}^{\infty}[\hat{A}_n r^{n-1}+\hat{B}_n r^{-n}]P_{n-1}(\cos\theta), \tag{12a}$$

$$T_\ell(r,\theta) = \sum_{n=1}^{\infty}[\hat{C}_n r^{n-1}+\hat{D}_n r^{-n}]P_{n-1}(\cos\theta), \tag{12b}$$

$$T_{\mathrm{g}}(r,\theta) = \sum_{n=1}^{\infty}[\hat{E}_n r^{n-1}+\hat{F}_n r^{-n}]P_{n-1}(\cos\theta). \tag{12c}$$

where $P_{n-1}(\cos\theta)$ is the $(n-1)$th-order Legendre polynomial.

Upon imposing the boundary conditions on the gas–liquid and liquid–solid interfaces from the continuities of the temperature fields and the heat fluxes and the boundedness requirement of T_{g}, one finds the six constants in (12) as

$$\hat{A}_n = \hat{T}_n N_n d^{n-1}[N_n-d^{2n-1}]^{-1}, \tag{13a}$$

$$\hat{B}_n = -\hat{A}_n/N_n, \tag{13b}$$

$$\hat{C}_n = -\frac{n-1+n\epsilon_1}{n-1+n\epsilon_1-(n-1)(1-\epsilon_1)b^{2n-1}}(N_n-b^{2n-1})\hat{B}_n, \tag{13c}$$

$$\hat{D}_n = -\frac{(n-1)(1-\epsilon_1)}{n-1+n\epsilon_1}\hat{C}_n, \tag{13d}$$

$$\hat{E}_n = \frac{(2n-1)\epsilon_1}{n-1+n\epsilon_1}\hat{C}_n, \tag{13e}$$

$$\hat{F}_n = 0, \tag{13f}$$

where

$$N_n = \frac{(n-1+n\epsilon_1)(n-1+n\epsilon_2)+n(n-1)(1-\epsilon_1)(1-\epsilon_2)b^{2n-1}}{(n-1)(1-\epsilon_1)[n+(n-1)\epsilon_2]+(n-1)(1-\epsilon_2)[n-1+n\epsilon_1]b^{-2n+1}},$$

$\epsilon_1 = k_\ell/k_g$, $\epsilon_2 = k_w/k_\ell$, k = thermal conductivity, $d = R_B/R_0$, and

$$\hat{T}_n = \tfrac{1}{2}(2n-1)\int_{-1}^{1} T_{ext}(\theta)\,P_{n-1}(\cos\theta)\,d\cos\theta,$$

where $T_{ext}(\theta)$ is the external temperature field on the outer surface of the spherical container, and R_0 the outer radius of the spherical container.

By substituting (13e,f) into (12c) and setting $r = 1$, one obtains the temperature on the bubble surface as

$$T_g(r=1,\theta) = \sum_{n=1}^{\infty} \frac{(2n-1)\epsilon_1}{n-1+n\epsilon_1-(n-1)(1-\epsilon_1)b^{2n-1}}\frac{N_n-b^{2n-1}}{N_n-d^{2n-1}} \times d^{n-1}\hat{T}_n P_{n-1}(\cos\theta). \tag{14}$$

2.3. *Discussion*

Let the temperature field on the outer surface of the spherical container be specified as

$$T_{ext}(\theta) = T_0 + R_0 \sum_{n=1}^{\infty} t_n P_n(\cos\theta). \tag{15}$$

then, the steady-state migration velocity of the bubble U at the centre of the spherical container can be estimated from (10) and (14) in terms of $T_{ext}(\theta)$. By iteration, one is able to determine the particular value of $T_{ext}(\theta)$ with which U is zero. This is the condition for a stationary bubble. Explicitly, it is

$$I_2 = \frac{4R_B^2(\rho_\ell-\rho_g)g}{\mu_\ell V_{ref} K_2(2-5b^3+3b^5)}. \tag{16}$$

For a small temperature difference, the gas–liquid interfacial tensions of most liquids are very close to being a linear function of the temperature. Therefore, I_2 can be evaluated analytically, and the temperature gradient needed to hold a bubble stationary is

$$(t_1)_s = -\frac{2}{3}\frac{N_2-d^3}{N_2-b^3}\frac{[1+2\epsilon_1-(1-\epsilon_1)b^3]}{\epsilon_1\sigma K_2(2-5b^3+3b^5)}\frac{R_B(\rho_\ell-\rho_g)g}{\gamma_1}, \tag{17}$$

which is a constant. In (17), $\gamma_1 = |\partial\gamma_{g\ell}/\partial T|$ and the subscript s denotes the value of t_1 for a stationary bubble. When a bubble is immersed in an unbounded fluid, i.e. $d = b \to 0$, (17) becomes

$$(t_1)_s \xrightarrow[d=b\to 0]{} -\frac{1}{3}\frac{1+\sigma}{\sigma}\frac{1+2\epsilon_1}{\epsilon_1}\frac{R_B(\rho_\ell-\rho_g)g}{\gamma_1}. \tag{18}$$

Note that only a linear temperature gradient or the P_1-mode of the externally applied temperature field will maintain a stationary gas bubble.

In the derivation of the velocity fields, the pressure on the surface of the bubble was not considered as the necessary boundary condition. However, it is not difficult to show that, as long as the gradient of the gas–liquid interfacial tension remains in the P_1-mode, the pressure on the surface of the bubble is automatically balanced. The details of this pressure balance calculation are presented in the Appendix.

Equation (10) is quite general in the sense that results for many specific cases can be derived from it.

(i) When $I_2 = 0$, i.e. there is no interfacial tension gradient,

$$\bar{U} \to \frac{2}{3\mu_\ell}\left[\frac{R_B^2(\rho_\ell-\rho_g)\,g}{K_2}\right][3+2\sigma-3(1-\sigma)\,b^5]^{-1}. \tag{19}$$

This same expression was previously derived by Haberman & Sayre (1950).

(ii) When $b \to 0$, i.e. for an unbounded liquid medium,

$$\bar{U} \to \frac{2}{3\mu_\ell}[R_B^2(\rho_\ell-\rho_g)\,g(1+\sigma)-\tfrac{1}{2}\mu_\ell\, V_{ref}\, I_2]\,(3+2\sigma)^{-1}. \tag{20}$$

This expression was first derived by Young *et al.* (1959). (Note that there were some typographical errors in their original paper.)

(iii) When $I_2 = 0$ and $b \to 0$, i.e. for an unbounded, isothermal liquid medium,

$$\bar{U} \to \frac{2}{3\mu_\ell}[R_B^2(\rho_\ell-\rho_g)\,g]\frac{1+\sigma}{3+2\sigma}. \tag{21}$$

This is exactly the Hadamard–Rybczynski (1911) formula.

(iv) When $I_2 = 0$ and $\sigma \to 0$, i.e. for a solid sphere inside a concentric spherical liquid container,

$$\bar{U} \to \frac{2}{9\mu_\ell}[R_B^2(\rho_\ell-\rho_g)\,g]\,[1-\tfrac{9}{4}b+\tfrac{5}{2}b^3-\tfrac{9}{4}b^5+b^6]\,(1-b^5)^{-1}. \tag{22}$$

This agrees with the expression previously obtained by Cunningham (1910), Williams (1915) and Lee (1947).

(v) When $I_2 = 0$, $b \to 0$ and $\sigma \to 0$, i.e. for a solid sphere in an unbounded fluid medium,

$$\bar{U} \to \frac{2}{9\mu_\ell}R_B^2(\rho_\ell-\rho_g)\,g. \tag{23}$$

This is Stokes' (1850) law.

3. Numerical examples

To illustrate the utility of the results derived in the previous section, let us consider a spherical glass shell of 2 mm outer radius and 0.01 mm wall thickness, filled with silicone oil DC 200 (20CS) at a temperature of 20 °C. Assume that a small air bubble is introduced into the oil at the bottom of the shell using a hypodermic needle. Since the oil is quite viscous, the bubble will have attained its terminal velocity (steady state) by the time it reaches the centre of the shell, provided that the bubble were small enough. The steady-state migration velocity of the air bubble at the centre of the glass shell can now be calculated from (10) by setting $I_2 = 0$ since there is no surface-tension gradient owing to the uniform temperature. The bubble velocity versus the bubble radius is plotted in figure 2. For comparison, the migration velocity of a bubble of the same size immersed in an unbounded liquid is also plotted. It can be seen from figure 2 that the difference in the velocities of the bubble in these two

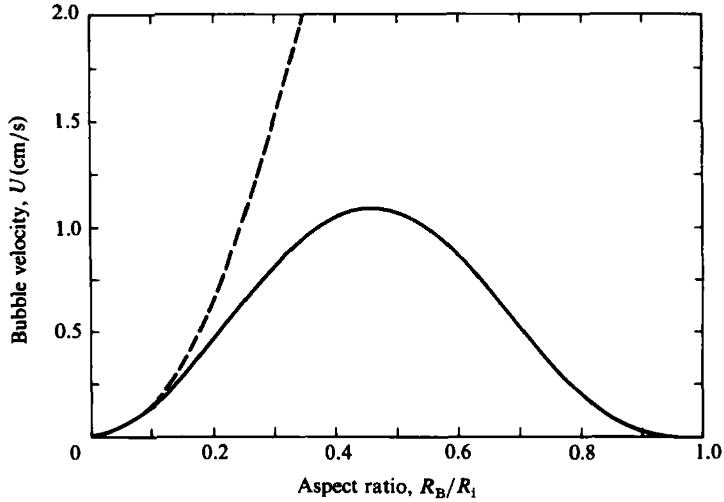

FIGURE 2. Velocity of an air bubble at the centre of an isothermal spherical glass shell. The outer radius and the wall thickness are, respectively, 2 mm and 0.01 mm. The shell is filled with silicone oil, DC200 (20CS) and is at a temperature of 20 °C. The broken line is the velocity of the bubble in an unbounded oil bath.

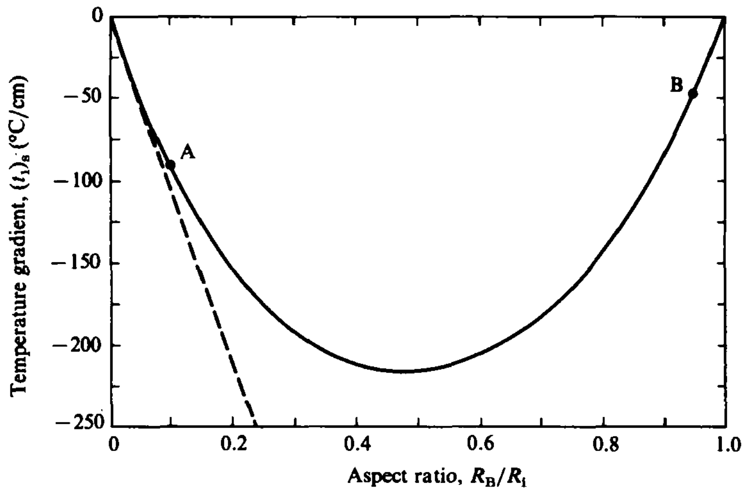

FIGURE 3. Temperature gradient on the outer surface of a spherical glass shell required to have a stationary air bubble concentric with the shell. The shell is filled with silicone oil, DC200 (20CS) and is at an average temperature of 20 °C. The outer radius and wall thickness of the shell are, respectively, 2 mm and 0.01 mm. The Péclet number is larger than unity between points A and B. The broken line is for a bubble in an unbounded oil bath.

different environments becomes significant when the aspect ratio, i.e. the ratio of the bubble radius to the container inner shell radius, R_B/R_i, is larger than 0.1.

The temperature gradient imposed on the outer surface of the glass shell that will maintain a stationary bubble at the centre of the glass shell is calculated from (17) and is plotted in figure 3. Note that the Reynolds numbers of the flows in the air and the oil are far below unity for all the bubble sizes considered. Therefore, the low-Reynolds-number approximation employed for the current work is justified. However, owing to the high Prandtl number of the oil, the Péclet numbers in the oil will be larger than unity when R_B/R_1 is between about 0.1 and 0.95, i.e. between points A and B marked in figure 3. In this range, the low-Péclet-number approximation of the heat equation will fail and a larger error will occur in the calculation of $(t_1)_s$.

When an air bubble is immersed in an unbounded oil bath, the temperature gradient in the oil that will keep the bubble stationary can be calculated from (18). In this case, the temperature gradient is a linear function of the bubble size, and is plotted in figure 3 for comparison. Because of the finite size of the glass shell, the temperature gradient $(t_1)_s$ is no longer a linear function of the bubble size when the bubble radius is larger than 0.04 mm. This effect of the finite size of the container on the temperature gradient that is required to maintain a stationary bubble is believed to be one of the reasons why the experimental data previously reported by Young *et al.* (1959) for larger-size bubbles are in poor agreement with the theoretical predictions.

4. Conclusions

The steady-state migration velocity of a gas bubble located at the centre of a spherical liquid bath has been calculated by considering the combined effect of gravity, interfacial tension gradient, and the finite size of the liquid container. A vertical temperature gradient will create an interfacial-tension gradient on the surface of the bubble and, hence, affect its vertical migration velocity. An analytical expression for the temperature gradient that will sustain a stationary bubble at the centre of the spherical container has been derived. The present result has also been compared with some of the earlier findings by others, giving more insight into the effect of the container wall on the bubble movement.

Finally, a comment is in order regarding the possibility of carrying out an analysis of the motion of the fluids, similar to the one presented in this work, when the gas–liquid interface is not concentric with the spherical container. Although the algebra required is certain to be much more involved and extremely tedious, such analysis is possible, and in fact quite straightforward, using the bipolar coordinates introduced earlier by Jeffery (1912). Calculations dealing with the non-concentric situation are currently in progress and will be reported in a future publication.

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Appendix

The pressure balance on the surface of a bubble located at the centre of a spherical container is calculated. The general expressions for the hydrodynamic pressures inside and outside the bubble are, respectively,

$$\tilde{p}_g = \text{Constant} - \mu_g V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} \left[\frac{2(2n+1)}{n-1} G_n r^{n-1}\right] P_{n-1}(\cos\theta), \tag{A 1}$$

$$\tilde{p}_\ell = \text{Constant} - \mu_\ell V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} \left[\frac{2(2n+1)}{n-1} C_n r^{n-1} + \frac{2(2n-3)}{n} D_n r^{-n}\right] P_{n-1}(\cos\theta). \tag{A 2}$$

The general expressions for the corresponding normal stresses are, respectively,

$$\tilde{\tau}_{rr,\,g} = \text{Constant} + 2\mu_g V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} [(n-2) E_n r^{n-3} + nG_n r^{n-1}] P_{n-1}(\cos\theta), \quad (\text{A } 3)$$

$$\tilde{\tau}_{rr,\,\ell} = \text{Constant} + 2\mu_\ell V_{\text{ref}} R_B^{-1} \sum_{n=2}^{\infty} [(n-2) A_n r^{n-3} - (n+1) B_n r^{-n-2} + nC_n r^{n-1} - (n-1) D_n r^{-n}] P_{n-1}(\cos\theta). \quad (\text{A } 4)$$

The pressure difference across the surface of the bubble is

$$\Delta\tilde{p}_{g\ell} = [\tilde{p}_\ell + \tilde{\tau}_{rr,\,\ell} - \tilde{p}_g - \tilde{\tau}_{rr,\,g}]_{\tilde{r}=R_B} - (\rho_\ell - \rho_g) g R_B \cos\theta + \text{Constant}. \quad (\text{A } 5)$$

Substituting (A 1), (A 2), (A 3) and (A 4) into (A 5), one has

$$\Delta\tilde{p}_{g\ell} = -I_2 \mu_g V_{\text{ref}} R_B^{-1} \cos\theta + \tfrac{1}{2}\mu_g V_{\text{ref}} R_B^{-1} \sum_{n=3}^{\infty} \frac{I_n}{n(n-1)(2n-1)} \times [\sigma K_n M_n - 6n] P_{n-1}(\cos\theta) + \text{Constant}, \quad (\text{A } 6)$$

where

$$M_n = 6 + n(4n^2-1)(n-2) b^{2n-3} - (2n+1)(2n-3)(2n^2-2n-1) b^{2n-1} + (2n-1)(2n-3)(n^2-1) b^{2n+1}.$$

The pressure-balance equation on the gas–liquid interface is the well-known Laplace–Young equation. For a spherical interface of radius R_B, it becomes

$$-\frac{2}{R_B}\gamma_{g\ell}(\theta) = \Delta\tilde{p}_{g\ell}. \quad (\text{A } 7)$$

The interfacial tension can be expanded into Legendre polynomials, namely

$$\gamma_{g\ell}(\theta) = \gamma_{g\ell,\,0} + \sum_{n=2}^{\infty} X_{n-1} P_{n-1}(\cos\theta), \quad (\text{A } 8)$$

where X_{n-1} is the expansion coefficient for the term containing P_{n-1}.

Substituting (A 6) and (A 8) into (A 7), one has

$$-\frac{2}{R_B}\gamma_{g\ell,\,0} - \frac{2}{R_B}\sum_{n=3}^{\infty} X_{n-1} P_{n-1}(\cos\theta) = \text{Constant} + \frac{2}{R_B}\sum_{n=3}^{\infty} \frac{X_{n-1} P_n(\cos\theta)}{4(2n-1)} [\sigma K_n M_n - 6n]. \quad (\text{A } 9)$$

From (A 9), it is clear that, for $n = 2$, namely when the interfacial tension is in the P_1-mode, the variation of the capillary pressure is completely counterbalanced by the pressure variation created by the fluid flows inside and outside the bubble. For $n > 2$, however, the capillary pressure variation cannot be counterbalanced since the following equality will never be satisfied:

$$\frac{\sigma K_n M_n - 6n}{4(2n-1)} = -1, \quad \text{for } n = 3, 4, 5, \ldots. \quad (\text{A } 10)$$

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