The motion of bubbles in a vertical temperature gradient

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It has been observed experimentally that small bubbles in pure liquids can be held stationary or driven downwards by means of a sufficiently strong negative temperature gradient in the vertical direction. This effect is demonstrated to be due to the stresses resulting from the thermal variation of surface tension at the bubble surface. The flow field within and around the bubble is derived, and an expression for the magnitude of the temperature gradient required to hold the bubble stationary is obtained. This expression is verified experimentally.

1. Introduction

It has long been known that when variations in temperature are maintained on the free surface of a pure liquid a dynamic steady state is achieved, characterized by a bulk flow in the liquid and at the surface, together with small surface deformations (Hershey 1939). The nature of this flow is as follows. A local increase in temperature results in a local decrease in surface free energy, γ; a surface temperature distribution therefore is accompanied by a non-uniform tangential stress in the surface, the positive direction of which is opposite to the surface temperature gradient. In response to this stress occurs a flow, the details of which are determined by a balance between the viscous shearing stress at the surface and the thermally induced surface stress. It is of interest to contrast the behaviour of a liquid subjected to a surface temperature variation with that of an elastic membrane subjected to the same thermal variation. Because the membrane is elastic, it may deform in its own plane until the tension is uniform throughout; the steady state in this case is therefore static, while in the case of the liquid a dynamic steady state is reached.

The situation in the liquid is entirely different if the surface is contaminated by an insoluble monolayer. Such monolayers can indeed support tangential stress without steady state flow by means of local density variations (Harkins 1952). This fact has been used as a very sensitive test for distinguishing between surface-tension induced flows, and flows due to other causes, e.g. free convection (Block 1956).

The direction of the flow in the surface is in the direction of the tangential stress vector. The direction of the surface flow is therefore away from the warmer

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regions, with liquid flowing to the surface from the bulk at the warm regions. Since the velocity does not vanish at the surface, a pressure variation is present at the surface. This variation in normal stress is balanced by a surface deformation; the excess normal stress necessary to balance the flow pressure is provided by the pressure due to surface curvature. Qualitatively, a depression in the surface is formed at a local hot spot, as has been shown by Hershey (1939).

In the course of a series of experiments on this phenomenon (other results of which will be published later), we have investigated the effect of temperature variations on a spherical free surface, e.g. that of a free bubble or droplet. According to the above argument, if the temperature increases monotonically from one pole to the other of a bubble surface, a flow must be produced if the liquid is uncontaminated. In the linear approximation to be described below, this flow is to be added to the normal Stokesian flow surrounding a freely rising bubble, so that the net effect must be to alter the motion of the bubble; this conclusion is, however, independent of whether the linear approximation is made.

It is easy to see in what direction the motion of the bubble will be altered. The capillary-type flow at the bubble surface, with respect to the bubble centre, is from the warmer to the cooler pole; with respect to the liquid bulk, therefore, the bubble will move in the direction of its warmer pole. Bubbles ought therefore to be 'attracted' by hot objects. This is easily demonstrated to be the case for small bubbles by touching a hot soldering iron to a test-tube containing any very clean liquid which has been shaken up to contain air bubbles.

2. Experiments and results

In order to avoid the effects of changing bubble solubility with temperature, the experiments were restricted to the determination of the relationship between the bubble radius and the (negative) vertical temperature gradient necessary to keep the bubble motionless with respect to the liquid at large distances. From dimensional analysis, this relationship may be expected to have the form

$$dT/dz = (K/\gamma')(\rho-\rho')gRF(\dots), \tag{1}$$

where dT/dz is the temperature gradient, K is a numerical constant, R is the bubble radius, γ' is the temperature coefficient of surface tension, ρ and ρ' are the densities outside and inside the bubble, and g is the acceleration of gravity. The function $F(\dots)$ is any function of dimensionless quantities, e.g. $(\mu/\rho)(gR^3)^{\frac{1}{2}}$, etc. The purpose of the experiment was to verify equation (1) and to determine the values of K and the function F.

The experiment was carried out by observing bubbles in a cylindrical sample of liquid carried in the gap between the anvils of a machinist's micrometer. The temperatures of the anvils were measured by means of mercury thermometers thrust into copper blocks borne by the anvils. The temperature of the lower block could be raised by increasing the current through a nichrome wire wrapped around the lower copper block. Measurements were confined to the region near the vertical axis of the liquid sample in order to minimize any effects associated with the free cylindrical surface. Bubble diameters were measured with a travelling microscope. The experimental arrangement is illustrated in figure 1.

In the experiment, small bubbles were found to collect at the lower (warmer) anvil. As the temperature gradient was slowly reduced, the larger of these were observed to detach and to rise slowly. A second adjustment of temperature gradient then made it possible to poise the bubble essentially motionless midway between the anvils. Measurements were made on all bubbles, the residual velocity being held by this means within the limits of $\pm 10\,\mu/s$. These bubbles were very nearly spherical in shape.

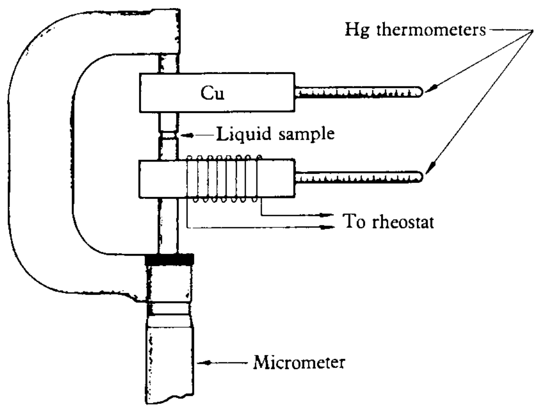

FIGURE 1. Schematic representation of experimental arrangement.

Fluid	ρ	$\mu \times 10^2$	γ	γ'* dyne cm^{-1} °C^{-1}	$\alpha \times 10^3$ deg^{-1}	$h \times 10^4$ cal/cm deg sec	c_p (cal/g)
n-hexadecane	0·773	3·52	27·66	−0·106	0·805	3·5	0·495
DC 200, 20 cs	0·955	19	20·50	−0·062	1·0	3·4	0·34
200 cs	0·971	193	21·04	−0·065	0·94	3·7	0·35
1000 cs	0·973	973	21·13	−0·061	0·94	3·8	0·37
Air, 20 °C, 1 atm.	0·0012	0·0018	—	—	—	0·57	0·25

ρ = density, μ = viscosity, γ = surface tension, γ' = thermal coefficient of surface tension, α = coefficient of volume expansion, h = thermal conductivity, c_p = heat capacity, c.g.s. units used throughout, temperature in °C.

* N. O. Young, 1955, *Rev. Sci. Inst.* **26**, 561. Probable error on γ' measurements is about 10 %.

TABLE 1. Fluid properties

In order to avoid free convective flow in the liquid sample, care was taken to keep the Rayleigh number, N, well below the critical value of about 2000.*

Measurements were carried out on air bubbles in the organic liquid n-hexadecane and in three Dow-Corning silicone oils of the DC 200 series. The viscosities of the three silicone oils differ widely while virtually all of their other specific properties are approximately the same. Specific parameters for each liquid are listed in table 1.

* The Rayleigh number is defined as $N = (\rho g a b^4/\mu\sigma)\,(dT/dz)$, where a, b, μ and σ represent the thermal coefficient of expansion, depth of the liquid, viscosity and thermal diffusivity, respectively (see Lin 1955).

The liquid *n*-hexadecane is easily contaminated by the introduction of very slight amounts of silicone oil, and was included for this reason. It was found that the introduction of very small amounts of silicone contaminant was sufficient to prevent completely the abnormal bubble behaviour under consideration, and this is taken as convincing evidence that the effect in question is a surface tension effect. On the other hand, it is known that it is very difficult to form monolayers on silicone, since virtually all contaminants remain in the bulk phase. In the present experiment it was found that the addition of relatively large amounts of contaminant to the silicone oils was ineffective in preventing abnormal motion.

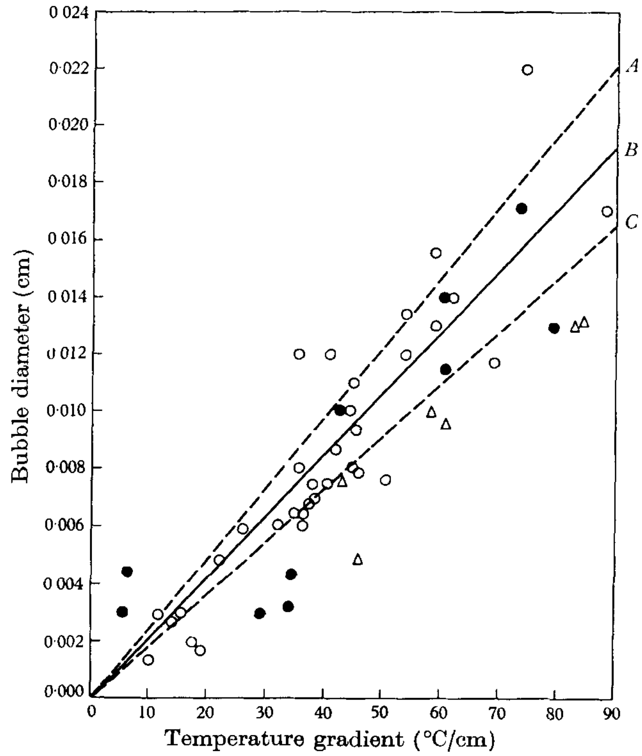

FIGURE 2. Bubble diameter, $2R$, *vs* temperature gradient necessary to hold bubble stationary. ○, DC 200, 20 cs; △, DC 200, 200 cs; , DC 200, 1000 cs.
$A = \gamma' = -0{\cdot}06$ dyne/cm; $B = \gamma' = -0{\cdot}07$ dyne/cm; $C = \gamma' = -0{\cdot}08$ dyne/cm.

The data for the three silicone oils is presented graphically in figure 2. The theory in the following section predicts that the value of K is $\frac{2}{3}$ and that the function F of equation (1) is equal to unity. Thus, the straight lines represent equation (1) (with $K = \frac{2}{3}$ and $F = 1$) for three different values of γ'. There are three principle sources of error involved in this determination; first, an uncertainty of about 10 % in the value of γ'; an estimated probable error of about 10 % in measuring bubble diameter (this error may perhaps be as large as 20–25 % for the smallest bubbles); and finally, an error resulting from the $\pm 10\,\mu$/s uncertainty in residual velocity. This error is greatest for those bubbles whose normal (Stokesian) velocity is smallest; thus, the residual velocity errors are greatest for the smallest bubbles, and for the highest viscosity.

It will be seen that the data confirm the theory within the limits of experimental accuracy. The critical temperature gradient is proportional to the bubble radius and is independent of viscosity (which varies by a factor of fifty among the three oils).

3. Derivation of the motion of a bubble in a vertical temperature gradient

In this section the flow field around a stationary undeformed bubble in a temperature field is derived. The analysis which follows is essentially a perturbation calculation, the unperturbed solution being one of no flow at all. It is then consistent with perturbation theory to neglect bubble deformations to the first order.

The centre of the bubble is taken as the origin of co-ordinates, and the z direction (direction of gravity) as the polar axis. Let ρ and μ be the density and viscosity, respectively, of the fluid outside the bubble (assumed to be infinite in extent) and let T be the temperature field outside the bubble. Corresponding quantities inside the bubble are denoted by the same symbols primed, e.g. ρ', μ', T'. The linearized Navier–Stokes equations describing the flow are (Birkhoff 1950)

$$\mu\nabla^2\mathbf{u} = \operatorname{grad}(p+\rho g z), \quad \operatorname{div}\mathbf{u} = 0, \tag{2}$$

where $\mathbf{u}$ is the flow field outside the bubble and p is the pressure. Identical equations describe the flow field $\mathbf{u}'$ inside the bubble. Quadratic (inertial) terms have been neglected in equation (2); this is the approximation of 'creeping flow' and may be expected to be valid for small bubbles under the same conditions which make valid Stokes's law for a freely rising bubble (Lamb 1945).

The energy equation governs the temperature distribution. If σ is the thermal diffusivity of the liquid outside the bubble, this equation has the form, for the steady state (Jakob 1942)

$$(\mathbf{u}\,.\operatorname{grad})\,T = \sigma\nabla^2 T. \tag{3}$$

A similar equation holds inside the bubbles for T'. If σ is sufficiently large, it is consistent with the approximation of creeping flow to neglect forced convection in determining the temperature distribution. With this approximation, T becomes a solution of Laplace's equation

$$\nabla^2 T = 0. \tag{4}$$

The axially symmetric solutions for flow around and within a spherical bubble have been derived by Rybczynski (1911) and by Hadamard (1911). If u_r and u_θ represent the radial and transverse components of the flow field outside the bubble, with u'_r and u'_θ their counterparts within the bubble, the solution is

$$u_r = [(a/\mu)\,(r^{-1}-R^2r^{-3})+v_0(1-R^3r^{-3})]\cos\theta, \tag{5a}$$

$$u_\theta = -[(a/\mu)\,(r^{-1}+R^2r^{-3})+v_0(1+\tfrac{1}{2}R^3r^{-3})]\sin\theta, \tag{5b}$$

$$u'_r = (a'/10\mu')\,(r^2-R^2)\cos\theta, \tag{5c}$$

$$u'_\theta = -(a'/10\mu')\,(2r^2-R^2)\sin\theta, \tag{5d}$$

$$p = (a/r^2)\cos\theta - \rho g r\cos\theta, \tag{5e}$$

$$p' = a'r\cos\theta - \rho' g r\cos\theta + a'_0, \tag{5f}$$

where a, a' and a_0' are constants to be determined by the boundary conditions, R is the bubble radius and v_0 is the velocity of rise of the bubble (later to be set equal to zero).*

We must include with these results the appropriate solutions of equation (4)

$$T = T_0 + T_1(r + k/r^2)\cos\theta, \tag{6a}$$

$$T = T_0 + T_1 k' r \cos\theta, \tag{6b}$$

where k, k', T_0 and T_1 are constants also to be determined from the boundary conditions.

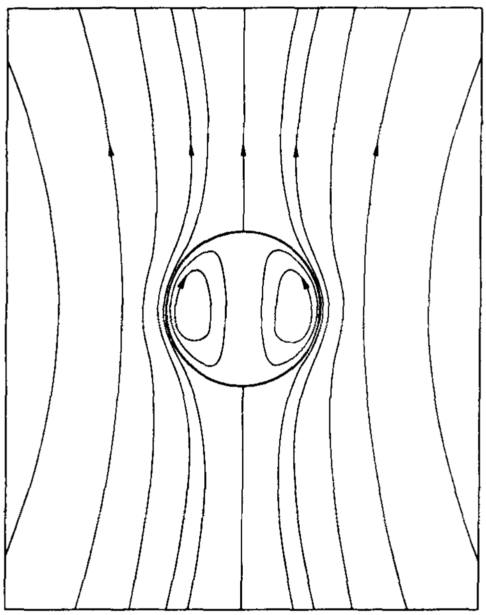

FIGURE 3. Flow field within and around the bubble.

The solutions expressed by equations (5) and (6) automatically satisfy conditions of regularity at the origin, and also satisfy the asymptotic conditions

$$\mathbf{u} \to (0, 0, v_0), \quad p \to -\rho g z, \quad T \to T_0 + T_1 z, \quad \text{as} \quad |\mathbf{r}| \to \infty. \tag{7}$$

Furthermore, the constants in these solutions have been chosen so that at the bubble surface $u_r = u_r' = 0$, since there can be no flow across the bubble surface.

Additional boundary conditions must be applied at $r = R$ to express (a) continuity of u_θ across the bubble surface, (b) continuity of shear stress across the surface, including that due to the thermal variation in surface tension, (c) continuity of normal stress across the surface, (d) continuity of temperature across the surface, and (e) continuity of heat flux across the surface. These conditions are expressed by

$$u_\theta(R, \theta) = u_\theta'(R, \theta), \tag{8a}$$

$$[\mu'(\partial u_\theta'/\partial r - u_\theta'/r) - \mu(\partial u_\theta/\partial r - u_\theta/r) - r^{-1}\partial\gamma/\partial\theta]_{r=R} = 0, \tag{8b}$$

$$[p' - p + 2\mu\, \partial u_r/\partial r - 2\mu'\, \partial u_r'/\partial r - 2\gamma/R]_{r=R} = 0, \tag{8c}$$

$$T(R, \theta) = T'(R, \theta), \tag{8d}$$

$$[h\, \partial T/\partial r - h'\, \partial T'/\partial r]_{r=R} = 0, \tag{8e}$$

* The solutions correctly describe the physical situation only when $v_0 = 0$.

where h and h' are the thermal conductivities outside and inside the bubble, respectively. When these five conditions are imposed on the solutions given by equations (7) and (8), the constants a, a', a_0, k and k' may be eliminated, and the single relation which obtains among the remaining parameters is

$$v_0 = (\tfrac{2}{3}\mu)\,[\mu\gamma' RT_c - (\rho-\rho')\,gR^2(\mu+\mu')]\,(3\mu+2\mu')^{-1}, \tag{9}$$

where $T_c = 3T_1/(2+h'/h)$. Setting $v_0 = 0$ in this equation gives the required relationship between the fluid parameters and dT/dz for the stationary bubble. Putting $\rho' = \mu' = h' = 0$, which is correct to within a few percent, yields equation (1) with $K = \frac{2}{3}$ and $F = 1$. The streamlines for the flow within and around the bubble are illustrated in figure 3.

Equation (9), it may be noted, agrees with the law derived by Hadamard and Rybczynski when $\gamma' = 0$. The term involving γ' enters in exactly the same way as the slip coefficient applied by Epstein (1924) to Stokes's law.

If in equation (9) we neglect unprimed quantities compared to primed ones, the case of the droplet falling in a gas results. It is seen that, by neglecting h and μ in equation (9), the term involving γ' disappears and Stokes's law is obtained. Thus the effect is extremely small for the case of droplets falling in a gas unless the droplet radius is of the order of $3\mu\gamma' hT_1/\mu' h'\rho' g$. It is possible that this situation is realized in the case of burning fuel droplets.

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REFERENCES

Birkhoff, G. 1950 *Hydrodynamics*. Princeton University Press.
Block, M. J. 1956 *Nature, Lond.*, **178**, 650.
Epstein, J. 1924 *Phys. Rev.* **23**, 710.
Hadamard, J. 1911 *C.R. Acad. Sci., Paris*, **152**, 1735.
Harkins, W. 1952 *The Physical Chemistry of Surface Films*. New York: Reinhold.
Hershey, A. V. 1939 *Phys. Rev.* **56**, 204.
Jakob, M. 1942 *Heat Transfer*. New York: Wiley.
Lamb, H. 1945 *Hydrodynamics*. New York: Dover.
Lin, C. C. 1955 *Hydrodynamic Stability*. Cambridge University Press.
Rybczynski, D. 1911 *Bull. Acad. Sci. Cracovie*, **1**, 40.