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Bubble shapes in rotating two-phase fluid systems: a thermodynamic approach

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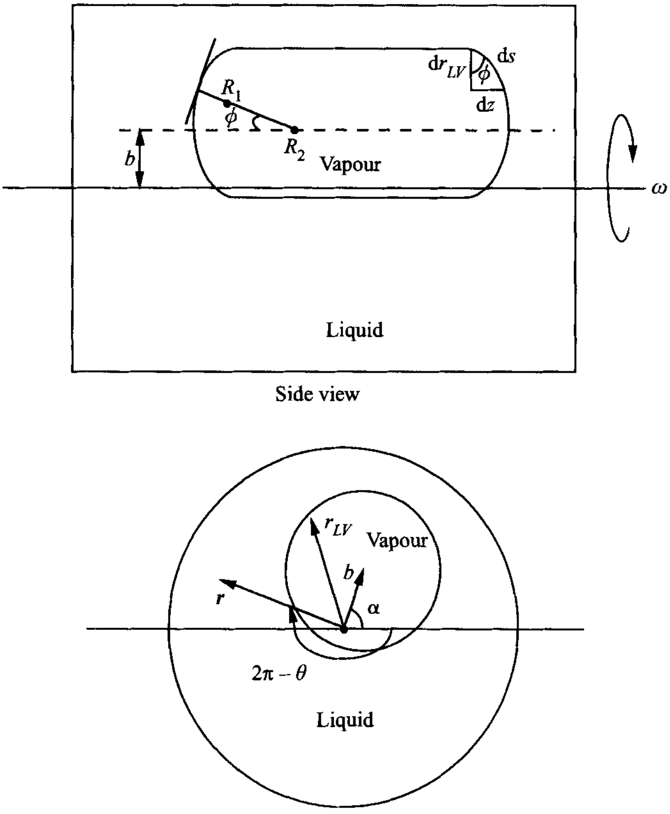
A method is reported for predicting the shape of the phase boundary in two-phase isothermal constant-volume constant-mass rotating fluid systems. In contrast to previous methods that have employed the continuum concept of pressure, the proposed method uses the thermodynamic concept. The latter requires, in addition to the usual condition of a force balance existing at the boundary, that the equilibrium phase boundary shape be such that there is no net mass flux. The latter condition is imposed by requiring that the chemical potentials in the different phases be equal at the phase boundary. A non-dimensional parameter is defined that allows one to determine when the effects of a gravitational field acting at 90° to the axis of rotation may be neglected. Experiments have been performed under conditions where this restriction is satisfied. With known values of the experimentally controllable variables, the proposed method has been used to predict the length of the vapour phase. To within the experimental error, the predicted lengths are found to be in agreement with the measurements. If, however, a gravitational field of a sufficient magnitude is imposed the vapour phase has been found to become unstable and to break into two or more separate bubbles. Using the variable-gravity environment of an aircraft following a parabolic flight path, this instability has been investigated. By approximating the gravitational effects, the theoretical description has been extended and a method proposed to determine the conditions under which the phase boundary becomes unstable. If the angle of action of the net viscous shear force on the bubble were known, a prediction of the breakup could be made entirely in terms of experimentally controllable parameters. Using arguments for the value of this angle, bounds on the breakup condition are compared with experimental results.

1. Introduction

The shape of the phase boundary in rotating liquid–vapour systems is important for applications such as the interfacial tensiometer (Cayias, Schechter & Wade 1975; Currie & Van Nieuwkoop 1982; Manning & Scriven 1977; Princen, Zia & Mason 1967; Torza 1975; Vonnegut 1942) and microgravity materials processing. Although the gravity level in the latter is small compared to that on Earth, it is not zero (Alexander 1990), and it is often necessary to determine its importance compared to that of rotational effects or of surface tension effects. Also, for such systems, instabilities have been observed to develop when the magnitude of the residual gravitational intensity bears a certain relation to the rotation rate (Yee, Wade & Ward 1991).

To investigate the source of these instabilities, we found it necessary to develop a new method for predicting the phase boundary shape when the two phases are present in



Side view

End view

FIGURE 1. Schematic representation of the system considered and of the coordinate systems used. The turning angle ϕ is defined as indicated. The two independent radii of curvature at any point on the phase boundary are defined by R_1 and R_2. The origin of the cylindrical coordinate system is taken as the position where the phase boundary crosses the axis of rotation of the cylinder.

an isothermal constant-volume system that is closed to mass transport and is being rotated. This method concentrates on making the prediction of the stable equilibrium shape in terms of the experimentally controllable variables for the problem. If gravitational effects are negligible these variables are the temperature, the total system volume, the total number of moles of the fluid and the rotation rate (i.e. T, V, N and ω respectively).

A related problem has been considered by a number of authors (Cayias *et al.* 1975; Princen *et al.* 1967; Rosenthal 1962) although their objective was somewhat different. Under the condition of negligible gravitational effects, their aim was to predict the equilibrium shape of the phase boundary in terms of the surface tension and other variables. Then the surface tension of the phase boundary was to be determined by fitting the predicted shape to that measured. However, the methods previously used included only one of the two necessary conditions for thermodynamic equilibrium. As a result, the predictions were not in terms of experimentally controllable variables, but in terms of empirical quantities that could not be determined *a priori* such as the bubble length (Cayias *et al.* 1975; Princen *et al.* 1967) or volume (Princen *et al.* 1967) or width (Rosenthal 1962).

The equilibrium condition included in previous analyses required that, at the phase boundary, a balance exist between the surface tension forces and those that arise from the pressures in the two fluid phases (Cayias *et al.* 1975; Princen *et al.* 1967; Rosenthal 1962; Rayleigh 1914). The equilibrium condition not imposed in previous analyses is the one that requires that the boundary shape be such that there is no net molecular transport across the phase boundary. This latter condition can be imposed by requiring the (electro)chemical potentials at the phase boundary to be equal (Gibbs 1961). This condition leads to a second restriction on phase boundary shape, i.e. to the Gibbs–Kelvin (Gibbs 1961; Thomson 1871) equation.

In the method we present for predicting the phase boundary shape, both conditions for equilibrium are imposed. The second equilibrium condition provides an additional equation and allows us to predict the phase boundary shape in terms of the experimentally controllable variables. The formulation of the equations leads to a condition for predicting when gravitational effects may be neglected, and since this condition may be written in terms of experimentally controllable variables, it may be quantitatively assessed. The validity of this new method is then examined by comparing the resulting predictions with a set of experiments that were conducted with a liquid of known surface tension and under conditions for which gravitational effects were negligible.

Under conditions where the gravitational effects are not negligible, an instability may develop that causes the vapour phase to become unstable and break into two or more separate bubbles (Yee *et al.* 1991). This instability develops when an acceleration (or gravitational intensity) is imposed in a direction perpendicular to the axis of rotation. The rippling of the phase boundary has been previously noted by other researchers (Princen *et al.* 1967; Vonnegut 1942), but not satisfactorily explained.

Pedley (1967) and Rosenthal (1962) have done theoretical work on fluid instabilities in systems that resemble the one being studied here, but for axisymmetric geometries with no gravitational forces present. The instability that we consider is only present when gravitational effects are present. Gans (1977) and Phillips (1960) have studied the stability of a partially filled rotating cylinder subjected to a gravitational acceleration perpendicular to the axis of rotation. In these studies, the lighter phase extended to the end of the cylindrical container so that the flow was essentially two-dimensional. The two-dimensional nature of the flow led to the vapour bubble being displaced *downward* from the axis of rotation.

By contrast, for the circumstances we consider, the vapour phase does not extend the full length of the container, giving rise to flow around the ends of the vapour phase and to a three-dimensional flow pattern. In this case, the effect of a gravitational field is to displace the bubble *upward* from the axis of rotation. By approximating the effects of the gravitational field, we present a prediction of the conditions under which the breakup of the vapour phase occurs. By assuming a direction of the viscous shear, the method presented for predicting the shape of the phase boundary entirely from experimentally controllable parameters allows us to evaluate the predicted breakup condition and compare the predictions with experimental results.

2. The governing equations

We consider a single-component two-phase rotating system that has constant volume and mass. Such a system is shown schematically in figure 1. The gravitational field acts perpendicular to the axis of rotation causing the bubble to be displaced from this axis by an amount b. For a system having given values of T, V, N and ω and a gravitational acceleration g, the thermodynamic potential is the Helmholtz function with the energy term augmented to take into account the potential energy arising from both the gravitational and centrifugal fields (Münster 1970). The Helmholtz function per unit volume in phase j is given by

$$f^j = (u^j + \chi^j) - Ts^j, \tag{1}$$

where u^j, χ^j and s^j are the internal energy, the potential energy arising from the fields and the entropy respectively, each per unit volume. To obtain the expression for the potential energy per unit volume, χ^j, we first introduce ψ, the potential energy of a unit volume fluid element per unit mass. If the position of a fluid element is denoted by $\boldsymbol{r}(r, \theta, z)$ and its acceleration by $\ddot{\boldsymbol{r}}$, then the potential per unit mass is given by (Münster 1970)

$$\psi = gr\sin\theta - \int_0^r \ddot{\boldsymbol{r}}\cdot \mathrm{d}\boldsymbol{r}, \tag{2}$$

where we have defined the location of zero potential to be the axis of rotation. If n^j is the number of moles per unit volume in phase j and W is the molecular weight, then the potential energy per unit volume in that phase is given by

$$\chi^j = n^j W\psi. \tag{3}$$

We shall adopt the Gibbs dividing surface approximation and treat the liquid–vapour interphase as a surface. Further, we shall assume that the position of the interphase is chosen such that the surface tension, γ^{LV}, does not depend explicitly on curvature.

To determine the necessary conditions for equilibrium, we suppose that the system has reached equilibrium and require that the variation of F (the total Helmholtz function) must vanish for all virtual displacements about the equilibrium state. This leads to the following two relations as the necessary conditions for equilibrium (Münster 1970):

$$\mu^j + W\psi = \xi, \quad j = L,\ V \text{ or } LV, \tag{4}$$

where μ^j is the chemical potential of the component in phase j and ξ is a constant; and

$$P^V_{LV} - P^L_{LV} = \gamma^{LV}[1/R_1 + (1/R_2)], \tag{5}$$

where a superscript L, V or LV on a property associates it with the liquid, vapour, or interphase respectively. The pressure is denoted as P, and R_1 and R_2 are the two independent radii of curvature at a point on the phase boundary that define the shape of the interphase. The subscript LV on a quantity indicates that it is to be evaluated at the liquid–vapour interphase. It is (4) that has been neglected in the past.

When (4) is applied at the phase boundary, it reduces to

$$\mu^L_{LV} = \mu^{LV} = \mu^V_{LV}. \tag{6}$$

Equations (4), (5) and (6) are the basic equations required to describe the equilibrium phase boundary shape. However, as written, they are too general to be readily applied. To write them in a more tractable form, we first consider the approximate expressions for the chemical potentials in each phase.

We shall approximate the liquid phase as slightly compressible so that, by definition, its isothermal compressibility, κ, is constant. Then if v^L is the specific volume of the liquid, one finds (Ward *et al.* 1983)

$$P^L = P^L_0 - \kappa^{-1}\ln(v^L/v^L_\infty), \tag{7}$$

where the subscript ∞ on a property indicates that it is to be evaluated at the saturated condition corresponding to the temperature. The Gibbs–Duhem relation for a bulk phase requires

$$n^j\,\mathrm{d}\mu^j = -s^j\,\mathrm{d}T + \mathrm{d}P^j, \quad j = L \text{ or } V. \tag{8}$$

After substituting (7) and (4) into (8), integrating under isothermal conditions and then simplifying, one finds

$$P^L = P_0^L - \kappa^{-1}\ln[1 + (\kappa W\psi/v_\infty^L)\exp\kappa(P_0^L - P_\infty)], \tag{9}$$

where the subscript 0 on a quantity indicates that it is to be evaluated on the axis of rotation. It should be noted that if the vapour phase extended the entire length of the volume (Gans 1977; Phillips 1960), this condition could not be applied.

We further restrict the system to cases for which the following are satisfied:

$$|\kappa(P_0^L - P_\infty)| \ll 1 \tag{10}$$

and

$$|\kappa W\psi/v_\infty^L| \ll 1. \tag{11}$$

Basically these restrictions limit the rate of rotation to values where the effect of the compressibility of the liquid phase on the pressure is negligible. After making use of (10) and (11) in (9), one finds

$$P^L = P_0^L - W\psi/v_\infty^L. \tag{12}$$

This latter equation then defines how the pressure will vary in the liquid phase for limited rates of rotation. The chemical potential for the slightly compressible liquid phase may be obtained by substituting (7) into (8), integrating under isothermal conditions and evaluating the integration constant at the saturated state to obtain (Ward *et al.* 1983)

$$\mu^L(T, P^L) = \mu^L(T, P_\infty) + \kappa^{-1}(v_\infty^L - v^L). \tag{13}$$

Now we turn to the vapour phase. It will be approximated as an ideal gas so that (Ward *et al.* 1983)

$$\mu^V = \mu^V(T, P_\infty) + \bar{R}T\ln(P^V/P_\infty), \tag{14}$$

where $\bar{R}$ is the gas constant. From the ideal gas equation of state and (4) and (8), one finds

$$P^V = P_0^V \exp(-\psi/\bar{R}T), \tag{15}$$

where P_0^V is the pressure within the vapour phase on the axis of rotation. We note that this condition cannot be applied if there is no vapour phase on the axis of rotation. We shall suppose that the rate of rotation is limited to values such that

$$|\psi/\bar{R}T \ll 1|. \tag{16}$$

Then (15) may be written

$$P^V = P_0^V - W\psi/v_0^V. \tag{17}$$

If (12) and (17) are now applied at the phase boundary and then subtracted, one finds

$$P_{LV}^V - P_{LV}^L = P_0^V - P_0^L + \psi_{LV}\Delta\rho, \tag{18}$$

where ρ is density and $\Delta\rho$ is defined by

$$\Delta\rho \equiv \rho_\infty^L - \rho_0^V. \tag{19}$$

Equation (18) is one of the two fundamental equations that will be employed to predict the equilibrium shape of the phase boundary. The other is the boundary condition that is obtained from (5). It is convenient to express this latter condition in terms of the mean curvature, $L_{0\epsilon}$, at the point where the phase boundary crosses the axis of rotation. The subscript ϵ indicates that the mean curvature at this point is an equilibrium property of the phase boundary (see below):

$$\frac{2}{L_{0\epsilon}} \equiv \left(\frac{1}{R_{10}} + \frac{1}{R_{20}}\right). \tag{20}$$

Then from (5) and (20), one finds

$$L_{0\epsilon} = 2\gamma^{LV}/(P_0^V - P_0^L). \tag{21}$$

Since the chemical potentials must be equal on the phase boundary (see (6)), this latter relation may be simplified. From (13) and (14), one finds

$$P_0^V = P_\infty \exp\left[(1/\bar{R}T\kappa)(v_\infty^L - v_0^L)\right]. \tag{22}$$

After evaluating (7) where the axis of rotation crosses the phase boundary and combining the result with (21) and (22), one finds

$$L_{0\epsilon} = \frac{2\gamma^{LV}}{P_\infty \exp\left[(1/\bar{R}T\kappa)(v_\infty^L - v_0^L)\right] - P_\infty + \kappa \ln(v_0^L/v_\infty^L)}. \tag{23}$$

Thus, the equality of the chemical potentials at the phase boundary leads to a condition that the equilibrium phase boundary shape must satisfy: its mean radius of curvature at the axis of rotation is given by (23).

It should be noted that gravitational effects do not appear explicitly in (23) owing to the choice of the axis of rotation as the reference associated with zero potential energy. Gravitational effects do, however, appear explicitly in the other fundamental equation that is available for predicting the phase boundary shape, (18). To see this dependence one may evaluate ψ_{LV} using (2) and insert the result in (18):

$$P_{LV}^V - P_{LV}^L = (2\gamma^{LV}/L_{0\epsilon}) + \Delta\rho g r_{LV} \sin\theta - \Delta\rho \int_0^{r_{LV}} \ddot{\boldsymbol{r}} \cdot \mathrm{d}\boldsymbol{r}. \tag{24}$$

Equation (24) may be used to predict the approximate phase boundary shape. It is not yet possible to predict the shape when both the centrifugal and gravitational fields are significant.

Note that in deriving the above equations, viscous effects were not considered. We shall consider two limiting cases. In one case, gravitational effects will be neglected completely. In this case, the system will be in rigid-body rotation and thus viscous effects will be absent. In the other case, gravitational effects will be taken into account approximately. In this case, the thermodynamic equation, (23), will not be solved, but rather an assumption about the bubble shape will be made. The Laplace equation will be used in the case with gravitational effects. The Laplace equation results from a mechanical force balance and as such is consistent with the presence of viscous effects. The effects of viscosity will be included through an additional force balance.

2.1. *Limit of negligible gravitational effects*

If gravitational effects are to be neglected, then the second term on the right-hand side of (24) must be negligible compared to the sum of the other two. This requires that the non-dimensional ratio Γ

$$\Gamma \equiv \left| \frac{g r_{LV} \Delta\rho \sin\theta}{(2\gamma^{LV}/L_{0\epsilon}) - \Delta\rho \displaystyle\int_0^{r_{LV}} \ddot{\boldsymbol{r}} \cdot \mathrm{d}\boldsymbol{r}} \right| \tag{25}$$

must be small compared to unity. To determine the magnitude of Γ, we suppose that the flow field may be approximated as purely rotational. Then

$$\ddot{\boldsymbol{r}} = r\omega^2 \boldsymbol{i}_r, \tag{26}$$

where $\boldsymbol{i}$ is a unit vector in the radial direction. Also, we introduce two non-dimensional parameters. One parameter, λ, compares the magnitude of the gravitational forces to the surface forces and the other, δ, compares the centrifugal forces to the surface forces:

$$\lambda \equiv gL_{0\epsilon}^{2}\Delta\rho/\gamma^{LV}, \tag{27}$$

$$\delta \equiv \omega^{2}L_{0\epsilon}^{3}\Delta\rho/\gamma^{LV}. \tag{28}$$

Then Γ may be written

$$\Gamma = \left|\frac{\lambda r_{LV}^{*}\sin\theta}{2-\delta r_{LV}^{*2}}\right|, \tag{29}$$

where r_{LV}^{*} is the position of the phase boundary non-dimensionalized with respect to $L_{0\epsilon}$. After requiring Γ to be small in (24), combining with (5) and non-dimensionalizing the result with respect to $L_{0\epsilon}$, one finds that the result may be written

$$\left(\frac{1}{R_{1}^{*}}+\frac{1}{R_{2}^{*}}\right) = 2-\delta r_{LV}^{*}, \tag{30}$$

where the superscript $*$ on a quantity indicates that it has been non-dimensionalized.

If the rotation rate is sufficiently large that gravitational effects may be neglected, then the vapour phase will be symmetric about the axis of rotation. The axis of rotation then crosses the phase boundary at the apex of the vapour phase. The two radii of curvature then become equal, and $L_{0\epsilon}$ is equal to the radius of curvature at the apex of the vapour phase:

$$R_{10} = R_{20} = L_{o\epsilon}. \tag{31}$$

Following the work of Bashforth & Adams (1883), the shape may be parameterized in terms of the turning angle, ϕ, and the coordinates r_{LV} and z_{LV} (see figure 1). The coordinates r_{LV} and z_{LV} are related to the radii of curvature of the phase boundary and the turning angle:

$$r_{LV} = R_{2}\sin\phi, \tag{32}$$

$$\mathrm{d}r_{LV}/\mathrm{d}\phi = R_{1}\cos\phi, \tag{33}$$

$$\mathrm{d}z_{LV}/\mathrm{d}\phi = R_{1}\sin\phi. \tag{34}$$

After solving for R_2 from (32), substituting into (30), solving for R_1, substituting the result into (33) and (34) and non-dimensionalizing the length variables r_{LV} and z_{LV} with respect to $L_{0\epsilon}$, one finds

$$\frac{\mathrm{d}r_{LV}^{*}}{\mathrm{d}\phi} = \frac{r_{LV}^{*}\cos\phi}{2r_{LV}^{*}-\sin\phi-\delta r_{LV}^{*3}} \tag{35}$$

and

$$\frac{\mathrm{d}z_{LV}^{*}}{\mathrm{d}\phi} = \frac{r_{LV}^{*}\sin\phi}{2r_{LV}^{*}-\sin\phi-\delta r_{LV}^{*3}}. \tag{36}$$

Note that (35) and (36) are the same as those that one would derive if gravitational effects were neglected at the outset.

The approach adopted previously (Cayias *et al.* 1975; Princen *et al.* 1967; Rosenthal 1962) has been to solve (35) and (36) without reference to the condition imposed on the equilibrium shape by (23). It should be noted that if the previous approach is adopted only the 'non-dimensional' shape can be obtained from the solution of these equations because the solution depends on δ. The previous approach was then to introduce some measured aspect of the vapour phase, such as its length (Cayias *et al.* 1975; Princen *et al.* 1967) or volume (Princen *et al.* 1967) or width (Rosenthal 1962), and from this information infer the value of δ. If an empirical parameter is introduced then the solution will not be in terms of the experimentally controllable variables: T, V, N and

ω. In these previous methods one could not determine from a given set of experimental variables whether gravitational effects were negligible, since δ also appears in (29). If δ is predicted in terms of the experimentally controllable variables, both the dimensional phase boundary shape and the condition for neglecting gravitational effects can be evaluated.

2.1.1. *Equilibrium phase boundary shape in terms of experimentally controllable variables*

We now propose to incorporate the additional equilibrium condition given in (23) into the prediction procedure and show that this leads to a method for predicting the phase boundary shape in physical space in terms of the experimentally controllable variables. We shall assume the surface tension is known, and suppose it is desired to predict the phase boundary shape.

For given values of T, V, N and ω, δ depends only on $L_{0\epsilon}$. Once the value of δ is known one may solve (35) and (36) using a simple numerical technique such as a fourth-order Runge–Kutta procedure. To determine the value of $L_{0\epsilon}$ in terms of a given set of experimentally controllable variables, we shall adopt an iterative procedure. The approximations that have been introduced in (10), (11) and (16) amount to assuming that the specific volumes v^L and v^V are uniform within the respective bulk phases for the rotation rates considered. Thus, we may write

$$v_0^L = (V - V^V)/(N - N^V) \tag{37}$$

and

$$v_0^V = V^V/N^V. \tag{38}$$

From (22), (37), (38) and the ideal gas equation of state, one finds

$$N^V = (V^V P_\infty/\bar{R}T)\exp[(v_\infty^L - (V - V^V)/(N - N^V))/\kappa\bar{R}T]. \tag{39}$$

The volume of the vapour phase may be expressed as

$$V^V = \int_0^L \pi r_{LV}^2 \,\mathrm{d}z, \tag{40}$$

where L is the length of the vapour phase. It is more convenient to write V^V in terms of the turning angle, the non-dimensional shape and $L_{0\epsilon}$ as

$$V^V = \pi L_{0\epsilon}^3 \int_0^\pi r_{LV}^{*2}(2 - \delta r_{LV}^{*2} - \sin\phi/r_{LV}^*)^{-1} \sin\phi \,\mathrm{d}\phi. \tag{41}$$

The iterative procedure for calculating the shape of the phase boundary may now be stated. We shall hypothesize $L_{0\epsilon}$ to have the value of R_0, and then evaluate the hypothesis by calculating the value of $L_{0\epsilon}$ from (23) and comparing the two values. To calculate $L_{0\epsilon}$ from its assumed value, one first evaluates δ from (28) and the assumed value, R_0. This allows the phase boundary profile to be calculated numerically from (35) and (36), giving $r_{LV}(\phi; \delta)$ and $z_{LV}(\phi; \delta)$. These may be used in (41) to calculate V^V. One may then iteratively calculate N^V from (39), v_0^L from (37) and finally $L_{0\epsilon}$ from (23). If the calculated value of $L_{0\epsilon}$ does not agree with the value assumed, the hypothesis fails. One then assumes a new value of R_0 and repeats the procedure.

For negligible gravitational acceleration and given values of T, V, N and ω, a necessary condition for the vapour phase to be *inside* the surface of curvature is that the values of $L_{0\epsilon}$ be positive. If the value of $L_{0\epsilon}$ is such that the radii of curvature R_1 and R_2 are both positive at all points on the boundary, then the vapour phase is simply connected and forms a prolate spheroid. The possibility of $L_{0\epsilon}$ being positive and R_1

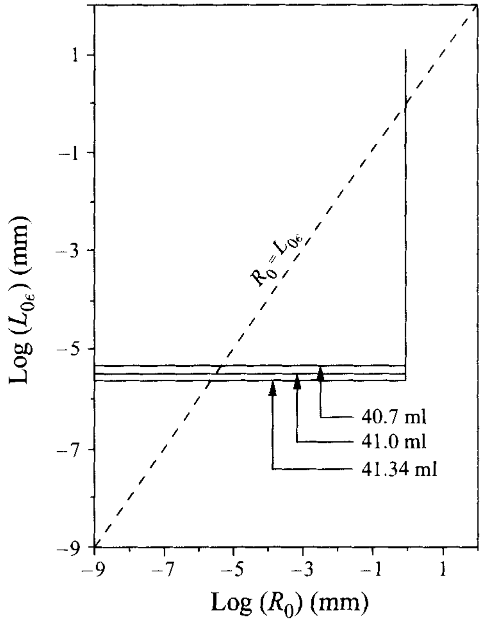


FIGURE 2. Determination of the possible equilibrium values of the radius of curvature of the phase boundary at its intersection with the axis of rotation. The equilibrium values are those for which the solid curves intersect the dashed line. The calculations are for three systems, each having a temperature of 20 °C, containing 40.05 g of H_2O, rotated at 50.97 Hz but having different volumes. For each set of experimentally controllable variables, there are two equilibrium radii.

or R_2 changing signs does not arise in this case because the pressure in the liquid phase would then be predicted to be dependent on z, and such a solution would be non-physical in the case under consideration. However, as will be seen, in the circumstance where gravitational effects are important, the possibility of $L_{0\epsilon}$ being positive and R_1 or R_2 changing signs does arise and limits the possible equilibrium shapes of the vapour phase.

For given values of T, N, ω and three different volumes, the solid curves in figure 2 show the values of $L_{0\epsilon}$ that are calculated by the procedure that was outlined. The dashed line indicates the positions where $L_{0\epsilon}$ is equal to R_0. As may be seen in this figure, for each set of experimentally controllable variables, there are two values of $L_{0\epsilon}$ for which the hypothesis is satisfied, i.e. at the intersections of the solid line with the dashed line. These are the values of $L_{0\epsilon}$ that satisfy all of the necessary conditions for equilibrium. We shall denote the smaller equilibrium value of $L_{0\epsilon}$ as L_{0e} and the larger as L_{0E}.

Since there are two possible equilibrium values of $L_{0\epsilon}$ for a given set of experimentally controllable variables, the question arises as to which of the possible equilibrium configurations the system will occupy. The situation of more than one equilibrium state has been encountered in a number of similar circumstances (Ward, Tikuisis & Venter 1982; Ward *et al.* 1983; Ward & Levart 1984; Ward, Yee & Tikuisis 1985) and in those circumstances the appearance of two equilibrium states could be physically understood once the thermodynamic stability of the different equilibrium states had been examined. The idea of such analyses is that equilibrium states correspond to extrema in the thermodynamic potential. The different types of extremum (i.e. maximum, local minimum or global minimum) correspond to different types of stability (i.e. unstable, metastable or stable equilibrium respectively). Following a similar approach to that

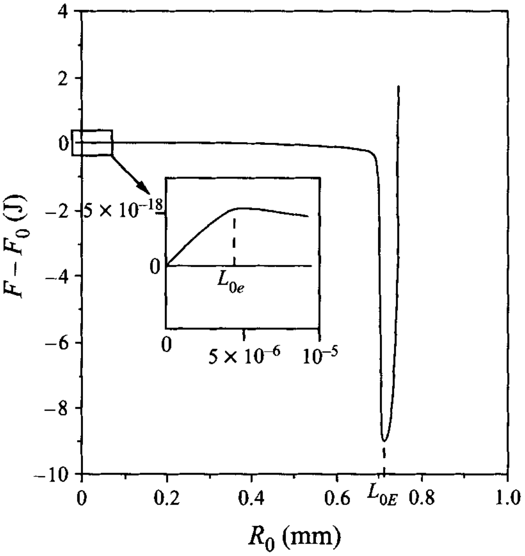


FIGURE 3. Examination of the thermodynamic stability of equilibrium sized bubbles. The Helmholtz function is shown as a function of the radius of curvature at the point where the phase boundary crosses the axis of rotation, R_0, for a system maintained at 20 °C, having a volume of 40.7 ml, containing 40.05 g of H_2O and rotated at 76.52 Hz. A bubble with the smaller value of $L_{0\epsilon}$ corresponds to a maximum in the Helmholtz function and is thus unstable, whereas a bubble with the larger value of $L_{0\epsilon}$ corresponds to a minimum of the Helmholtz function and is thus stable.

used by Ward *et al.* (1982, 1983, 1984, 1985), the difference in the Helmholtz potential from a reference state was evaluated for different values of R_0 and hence different configurations. The reference state that was used is that in which the system consists solely of a homogeneous liquid phase. The result of this procedure for a particular case is shown in figure 3.

The smaller equilibrium value of $L_{0\epsilon}$ (i.e. L_{0e}) corresponds to a maximum in the thermodynamic potential and is thus an unstable equilibrium size for the vapour phase, whereas the larger equilibrium value, L_{0E}, corresponds to a stable equilibrium size. Physically, if the liquid phase were initially homogeneous with the given values of T, V, N and ω, then L_{0e} would define the threshold size that must be produced in the nucleation event in order for homogeneous bubble nucleation to occur in the liquid phase. If a bubble of size greater than this were produced in the nucleation event, it would grow to the size corresponding to L_{0E} where it would be stable (Forest & Ward 1977, 1978; Tucker & Ward 1975; Ward, Balakrishnan & Hooper 1970; Ward *et al.* 1985). An example of the variation of the parameter L_{0E} with rotation rate when the other thermodynamic variables are fixed is shown in figure 4.

2.2. *Gravitational effects on phase stability*

We shall now consider the case where gravitational effects can no longer be neglected. When a rotating two-phase system such as that considered above is subjected to a gravitational intensity perpendicular to the axis of rotation, the system is no longer axisymmetric. The bubble moves off the axis of rotation and may change shape in response to forces arising from the gravitational field and the changing flow field. As the gravitational intensity is increased (for a fixed rotation rate) an instability has been observed to develop (Yee *et al.* 1991). Ripples grow on the phase boundary and at high enough gravitational intensities, the vapour phase breaks into two or more separate

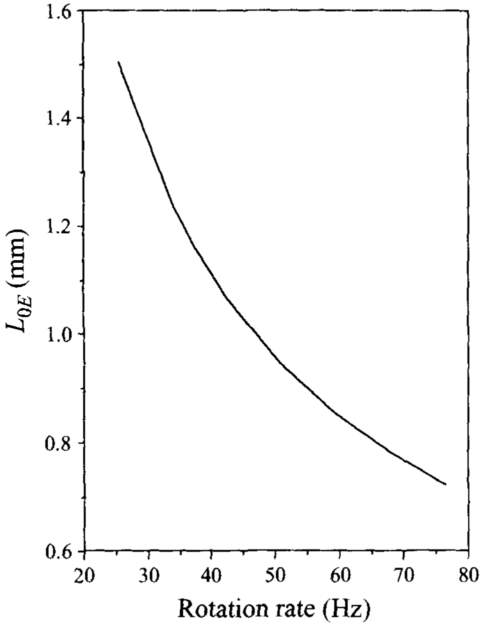


FIGURE 4. L_{0E} as a function of rotation rate for a system containing 40.05 g of H_2O in a volume of 40.7 ml with a temperature of 20 °C.

bubbles. One such breakup is shown in figure 5. A preliminary experimental study of this breakup phenomenon using water as the experimental fluid has been previously reported (Yee *et al.* 1991). Experiments (described in §3) show that for a fixed gravitational intensity, there is a lower limit to the rotation rate for which the vapour phase will remain as a single bubble. Also, for a fixed rotation rate, there is an upper limit to the gravitational intensity for which the vapour phase will remain as a single bubble.

We present here a theoretical prediction of the condition for breakup of the vapour phase. In order to arrive at an equation for the condition for breakup, gravitational effects will be included in an approximate way. We will assume that the sole effect of gravity is to move the bubble off the axis of rotation. We will assume that the bubble shape remains essentially the same as it was in the zero-gravity case. By this we mean that the bubble cross-section remains circular with the same radius as in the zero-gravity case and the same radius of curvature on the axis of rotation. We will also assume that the flow field remains centrifugal. These assumptions make quantitative calculations possible and as we will show, they yield physical insight into the breakup phenomenon.

As a possible criterion for the breakup of the vapour phase, we shall examine the condition under which zero mean curvature exists at some point on the phase boundary. Mean curvature can only be equal to zero if both curvatures at a point are equal to zero (the interphase is flat), or the two radii of curvature have opposite signs but are equal in magnitude. In either case, zero mean curvature is the condition that the interphase must pass through in order to change from the axisymmetric stable shape to a shape that has an 'indentation' and for which breakup is imminent. Such an indentation may be seen in figure 5 in the centre of the vapour phase before the bubble breaks into two portions. Thus we see that investigating points on the interphase where the mean curvature is equal to zero is identical to investigating points

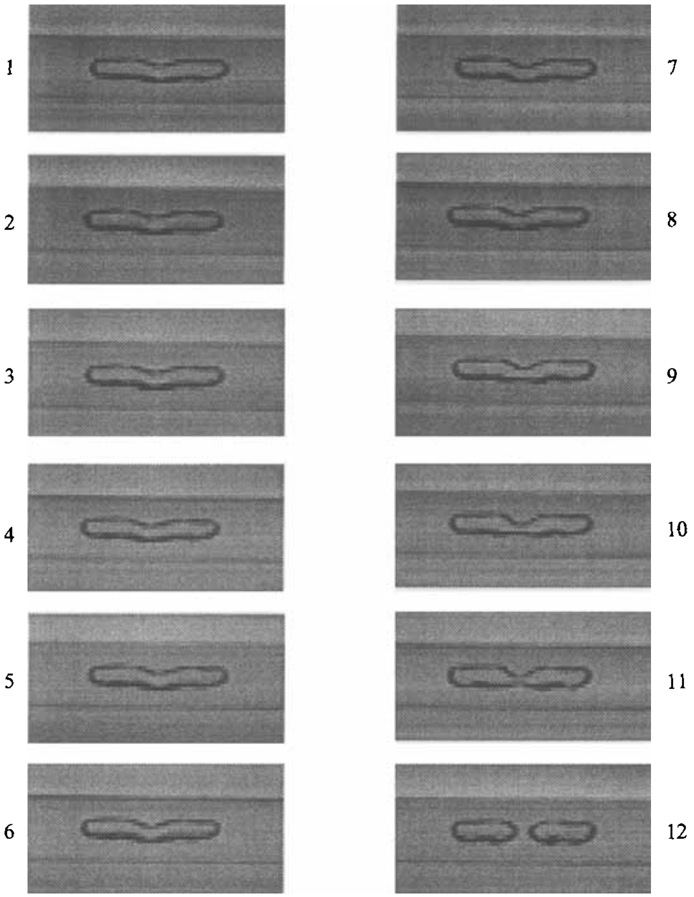


FIGURE 5. A sample breakup for 44.8 g of water in 45.89 ml at 36 °C rotating at 16.98 Hz. The time separation between successive images is 1/30 of a second. The acceleration changes from $0.788g_0$ for images 1–6 to $0.924g_0$ for images 7–12.

where an indentation of a certain size exists on the interphase. That certain size is such that the two orthogonal radii of curvature that define the system are of opposite sign but are equal in magnitude.

Ideally we would use the complete shape equation, (24), to find the condition under which zero mean curvature would exist. However, the three-dimensional nature of the equation, along with the fact that an iterative procedure must be used to obtain the solution, makes using the complete equation impractical. Hence, in order to calculate the condition for zero mean curvature at a point on the bubble surface, we will use the axisymmetric shape found by assuming that gravitational effects are negligible and assume that the effect of introducing gravity is to displace the bubble from the axis of rotation by an amount denoted by b (see figure 1). We assume that the pressure field remains centrifugal in order to be consistent with our approximation that the sole effect of gravity is to move the bubble in the field. The pressure profiles are then

$$P^L = P_0^L + Wr^2\omega^2/(2v_\infty^L) \tag{42}$$

and

$$P^V = P_0^V + Wr^2\omega^2/(2v_0^V). \tag{43}$$

Looking at the Laplace equation, (5), it is clear that zero mean curvature will exist at a point where the pressure in the vapour equals the pressure in the liquid. For the axisymmetric shape, the pressure in the vapour is greater than the pressure in the liquid. Since the specific volume of the liquid is smaller than that of the vapour, the pressure in the liquid increases with distance from the axis of rotation and the pressure in the vapour is comparatively unchanged. The point on the phase boundary to first reach a condition of zero mean curvature will be that point on the phase boundary where the liquid pressure is a maximum, i.e.

$$r_{LV} = r_{max} + b, \tag{44}$$

where r_{max} is the maximum radius of the bubble which can be found once the phase boundary profile has been calculated using the method described in §2.1.1. Thus the distance that the bubble may move off the axis of rotation before zero mean curvature can exist on the bubble surface may be found by requiring that the pressure in the vapour be equal to the pressure in the liquid at the point defined by (44):

$$P^V_{LV} - P^L_{LV} = P^V_0 - P^L_0 - \tfrac{1}{2}\Delta\rho(r_{max} + b)^2\omega^2 = 0. \tag{45}$$

Solving (45) for b and making use of (5) and (28) to eliminate pressure, yields as the displacement for which zero mean curvature occurs on the bubble surface (or in other words, breakup is predicted)

$$b = -r_{max} + L_{0\epsilon}(2/\delta)^{1/2}. \tag{46}$$

Note that the above equation was based on the Laplace equation with physical assumptions on the shape of the bubble in the presence of a gravitational intensity. Owing to the assumptions and the somewhat arbitrary selection of zero mean curvature as the condition for breakup, (46) will be qualitative only. In order to evaluate the gravitational intensity corresponding to the above condition from the experimentally controllable variables, a relation for b must be found.

2.2.1. *Prediction of the position of the bubble from knowledge of the flow field*

In order to determine the position of the bubble precisely, the viscous forces arising from the flow field must be found. The exact flow pattern around a displaced bubble is unknown. Currie & Van Nieuwkoop (1982) have predicted and observed the dominant flow patterns in different regions around a displaced bubble of circular cross-section. The forces acting on the bubble were calculated from these dominant flows (Currie & Van Nieuwkoop 1982).

There is a buoyancy force, F_B, acting vertically:

$$F_B = V^V(\rho^L - \rho^V)g. \tag{47}$$

The pressure field is assumed to remain centrifugal so that there is a centrifugal force, F_C, acting towards the axis of rotation of the cylinder:

$$F_C = \pi L r^2_{max} b \rho^L \omega^2, \tag{48}$$

where L is the length of the bubble. Note that the centrifugal force was calculated from the pressure difference around the portion of the volume that is in rigid-body rotation with the bubble (Currie & Van Nieuwkoop 1982). This portion of the volume is cylindrical with length equal to the length of the bubble and radius equal to the maximum radius of the bubble.

There are two viscous forces which Currie & Van Nieuwkoop (1982) took into account. The first is an Ekman force which is due to the Ekman layers arising at the ends of the bubble. Currie & Van Nieuwkoop have shown this force to be given by

$$F_E = 2\pi r^2_{max} b(\rho^L \omega^3 \eta)^{1/2}, \tag{49}$$

where η is the viscosity of the liquid. This force acts at an angle of 45° to the displacement vector. Since the Ekman force acts on a plane perpendicular to the axis of rotation and at a position where there is no viscous shear, it may only be balanced by the gravitational force which is purely vertical. If the Ekman force is to be balanced by the gravitational force, the only position for the bubble to be is at an angle of 45° to the horizontal. This angle is denoted α in figure 1. Currie & Van Nieuwkoop (1982) observed n-butanol drops in water to be displaced at an angle of 45°. We measured this angle approximately for a water vapour bubble rotating at three different rates in a large liquid volume (see §3.4) and obtained an angle of $44 \pm 1°$.

The second viscous force is that arising from viscous shear around the bubble. This force acts at an angle β to the horizontal. If the bubble is perfectly axisymmetric, Currie & Van Nieuwkoop have shown that the direction of the viscous shear is horizontal. Since they used small drops of n-butanol with volumes on the order of µl, the surface tension forces would be large enough to make axisymmetry a good assumption. In our case, the volume of the vapour is on the order of ml. Thus the weaker surface tension forces may allow the bubble to change its shape slightly, altering the angle of action of the viscous shear. The angle β will thus be left in the force balance.

A force balance, performed in a direction perpendicular to the viscous shear, requires

$$F_C \cos(45° - \beta) + F_E \cos(\beta) = F_B \cos(\beta). \tag{50}$$

Substituting (47), (48) and (49) into (50) yields as the displacement of the bubble

$$b = \frac{\cos\beta}{\cos(45° - \beta)} \frac{\rho^L - \rho^V}{\rho^L} \frac{V^V}{\pi L r_{max}^2} \left[1 + \frac{2}{L}\left(\frac{\eta}{\omega\rho^L}\right)^{1/2} \frac{\cos\beta}{\cos(45° - \beta)}\right]^{-1} \frac{g}{\omega^2}. \tag{51}$$

Substituting (51) into (46) and solving for g we get as the expression for the gravitational intensity at breakup, g_b

$$g_b = \frac{\cos(45° - \beta)}{\cos\beta} \frac{\rho^L}{\Delta\rho} \frac{\pi L r_{max}^2}{V^V} \left[1 + \frac{2}{L}\left(\frac{\eta}{\omega\rho^L}\right)^{1/2} \frac{\cos\beta}{\cos(45° - \beta)}\right]\left[-r_{max} + L_{0\epsilon}\left(\frac{2}{\delta}\right)^{1/2}\right]\omega^2. \tag{52}$$

Thus the gravitational intensity necessary to produce the condition of zero mean curvature somewhere on the bubble may be calculated from experimentally controllable variables and a value for β.

Arguments can be made for limits to the angle β and thus we may predict the approximate bounds on the zero-mean-curvature condition. Currie & Van Nieuwkoop (1982) have shown that if the bubble cross-section is circular, then the angle β is 0°. On the other hand, if the surface tension effects are not so large, the phase boundary can change its shape. The phase boundary cross-section will assume an airfoil-type shape, which will then orient itself in the direction of the flow. In this case the viscous shear will act in a direction closer to the flow direction and β will then be greater than 0°. In our study we have observed these airfoil-type shapes for large bubbles at low rotation rates.

2.2.2. *The limiting case of a long cylinder with low surface tension*

Under certain limiting circumstances, the breakup condition that has been derived can be shown to be similar to a previously obtained result. In the limit of large rotation rates the bubble becomes a very long cylinder (i.e. $L_{0E}/L \ll 1$). Princen *et al.* (1967) have shown that in this limit

$$\delta = \tfrac{16}{27} \tag{53}$$

and
$$L_{0E} = \tfrac{2}{3} r_{max}. \tag{54}$$

For a long cylinder, the approximate volume of the bubble is given by

$$V^V \approx \pi L r_{max}^2. \tag{55}$$

Also, if we consider the surface tension effects to be sufficiently small that the surface can change shape and orient itself in the flow, then

$$\beta \approx 45^\circ. \tag{56}$$

Making use of the limits given in (53)–(56) in (52), we get as the equation for the maximum gravitational intensity for stability

$$g_b = \sqrt{2}\frac{\rho^L}{\rho^L-\rho^V}\left[1+\frac{2}{L}\left(\frac{2\eta}{\omega\rho^L}\right)^{1/2}\right]\left(\left(\frac{3}{2}\right)^{1/2}-1\right) r_{max}\,\omega^2. \tag{57}$$

For the range of liquid viscosities and densities we shall consider, the second term in the square brackets is negligible compared to unity. Neglecting this term and neglecting the density of the vapour compared with the density of the liquid, we get

$$g_b = (\sqrt{3}-\sqrt{2})\, r_{max}\,\omega^2 \cong 0.318 r_{max}\,\omega^2. \tag{58}$$

Phillips (1960) gave a stability criterion for a rotating partially filled cylinder in which the air column extended to the ends of the cylindrical container. The phase boundary was considered to be a free surface and the stability criterion was found by perturbing the Euler equations. Gans (1977) extended the stability condition of Phillips to include some viscous effects. Using the notation developed here, Gans' condition is equivalent to (Greenspan 1975)

$$g_b = 0.314 r_{max}\,\omega^2. \tag{59}$$

The condition presented in (58) is for the stability of the vapour phase and was derived from a thermodynamic prediction of the shape of the bubble. Gans' condition, presented in (59), is for the stability of the liquid flow and was derived by perturbing the fluid mechanics equations of motion. As may be noted from (58) and (59), the conditions for stability predicted from these two approaches are very nearly the same. This would mean that the condition for the liquid to become unstable is the same as the condition for the vapour phase to break apart. The similarity of these two results is not surprising since: (i) the breakup condition was based on the Laplace equation (which is a force balance equation) along with physically reasonable assumptions about the shape of the displaced bubble; (ii) in (58) fluid mechanical forces were used to predict the position of the bubble; (iii) both the liquid flow and the vapour phase become unstable as the breakup occurs.

3. Experimental investigation

A series of experiments was performed to investigate the equations found in §2. A single apparatus was used to investigate both the phase boundary shape in rotating liquid–vapour systems in the absence of gravitational effects and the instability that develops when a gravitational field is added perpendicular to the axis of rotation. The apparatus was flown on board an aircraft (NASA's KC-135) that flew along a parabolic flight path to provide periods of low gravitational intensity (± 0.05 times standard gravity, g_0, for approximately 20 s at a time). Using the reduced gravity environment, the phase boundary shape predictions could be examined in a regime

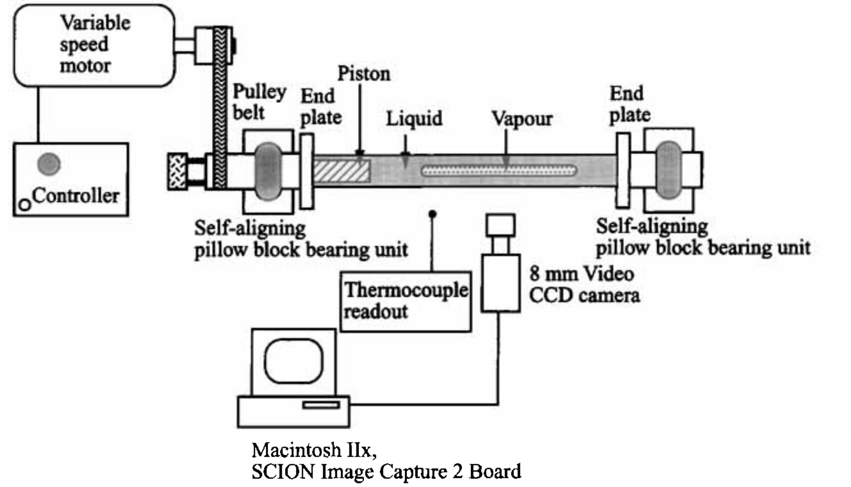


FIGURE 6. Schematic of the experimental apparatus used to examine the predictions.

where gravitational effects were negligible. Using the variable gravitational intensity produced as the aircraft flew multiple parabolas, the instability could be examined.

3.1. *Experimental apparatus for the study of vapour-phase shapes*

The experimental apparatus has been previously reported in Yee *et al.* (1991). A schematic diagram is shown in figure 6. A 250 mm long glass cylinder with an inner diameter of 16 mm and an outer diameter of 22 mm was first filled completely with liquid. The cylinder was closed by two end plates, one of which housed a threaded piston which could be retracted to produce a vapour bubble. To determine the mass of water present in the cylinder and its initial volume, the cylinder and end plates were weighed when filled and when empty. The volume change that resulted from rotating the threaded piston could be simply calculated using the pitch of the threads and the cross-sectional area of the piston. The cylinder could be rotated about its axis at speeds of up to 75 Hz. An error in the rotation rate of ± 0.083 Hz was assigned. This was the repeatability of the operator setting the speed in the adverse conditions of the parabolic flight. The temperature was recorded from a thermocouple placed outside the cylinder.

The phase boundary was photographed with a video camera that could be played back to examine the shape frame by frame. The images recorded from each experiment were stored and digitized. Since the apparatus was used in the variable gravity environment of the parabolic flight aircraft, the acceleration level was recorded using an accelerometer (Q-Flex model 1400 – Sundstrand Data Control Inc., Washington) with a readout that was within the view of the video camera.

The length of the vapour phase was first measured in units of pixels and then converted to millimetres. The pixel to millimetre conversion was found at the end of each experiment by recording the image of a scale when the scale was at the same position as the longitudinal axis of the vapour phase and when the video camera was at the same position as when the image of the vapour phase had been recorded for that particular experiment. The pixel to millimetre conversion introduces the largest error in bubble length measurements.

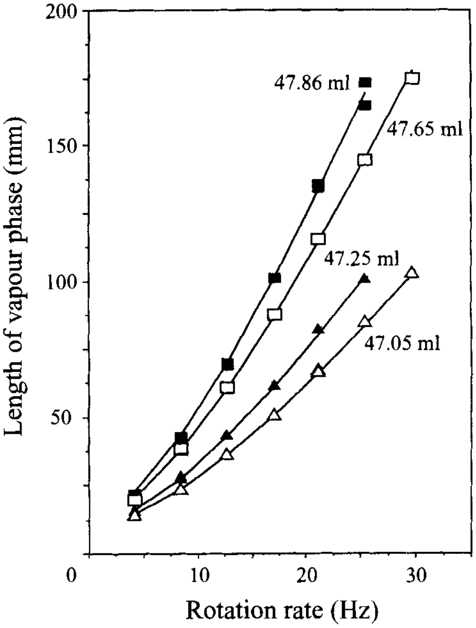


FIGURE 7. Comparison of the predicted lengths of the vapour phase with those measured for experiments performed at reduced gravitational intensities. The predictions are based on a nominal fluid mass of 38.0 g of octyl alcohol and the nominal volumes and rotation rates indicated. The temperatures used for each of the predictions were inferred from extra experiments at higher rotation rates (not shown here). The temperatures were 25.3 °C, 26.3 °C, 26.1 °C and 25.3 °C respectively starting from the smallest volume.

3.2. *Experimental procedure for examining the vapour-phase shapes in the absence of gravity*

To examine the predictions, we confine our attention to the stable equilibrium shapes of the vapour phase, i.e. those corresponding to L_{0E}. The experimental system was first filled with a given mass of octyl alcohol. The piston was then retracted to form four different system volumes for four series of experiments. In each series of experiments, the system volume, temperature and the number of moles were held constant. The experimental conditions are given in figure 7. For each experiment, the cylinder was first rotated at a rate that was sufficient for the length of the vapour phase to almost fill the cylinder. This was expected to bring all of the bubbles in the system to the longitudinal axis of the cylinder and to merge them into one vapour volume. The rotation rate was then set in the low gravity portion of the parabolic flight. The system had approximately 20–25 s to come to its equilibrium state before the onset of the increased gravitational intensity. The image of the vapour phase was recorded as was the rotation rate and the temperature. The gravitational intensity was monitored so that one could be sure that the image used for analysis was recorded at a time when the magnitude of the gravitational intensity was below $0.01g_0$.

On the aircraft, the temperature was not as stable as it would be in the laboratory. As a result, the temperature recorded using the thermocouple placed outside of the cylinder was not accurate enough. For each series of experiments, an extra experiment (at a higher rotation rate) was performed in order to infer the temperature using the theory. The temperature so inferred could then be used to independently examine the results of other experiments performed with the same volume and within a short time during which the temperature of the liquid remained approximately constant. The

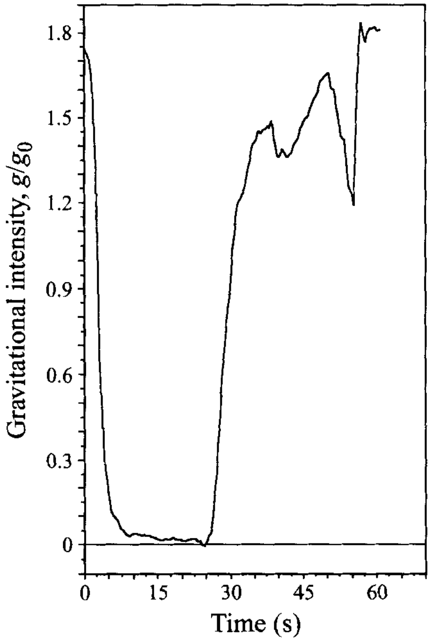


FIGURE 8. Gravitational intensity versus time for a particular parabola (parabola no. 19 on August 7, 1991).

differences between the temperatures measured outside the cylinder and the inferred temperatures ranged between 2.4 and 4.5 °C and were in a direction consistent with the hypothesis that the fluid temperature was lagging behind the changing temperature in the aircraft.

3.3. *Experimental examination of the breakup condition*

Once a vapour phase has been introduced into the rotating cylinder of the apparatus described in §3.1 above, the instability may be investigated in two ways. First, the gravitational intensity may be held fixed (at standard gravity in the laboratory) and the rotation rate decreased until breakup occurs. Conversely, the rotation rate may be fixed and the gravitational intensity increased (by taking the experiment on a parabolic flight) until the vapour phase breaks apart. We have used both procedures.

Experiments were first performed in the laboratory in which the rotation rate was incrementally reduced until the vapour phase broke apart. The limiting rotation rate was then taken as the average of the last setting before breakup and the setting that produced breakup. An error of half this interval was assigned. Care was taken to ensure that the system had reached its equilibrium state at one rotation rate setting before the rate was reduced to the next value. The limiting rotation rate appears to be independent of the rate at which the rotation speed is reduced. The time for which the apparatus was left at each setting during a particular experiment was varied from 0.5 to 60 min and the resulting limiting rotation rate remained the same.

In order to subject the rotating apparatus to a variation in the gravitational intensity, the apparatus was taken on board an aircraft that flew parabolas. An acceleration profile from one such parabola used for the variable gravity experiments is shown in figure 8. Initially the cylinder was rotating at a high enough rate to keep the vapour phase in one piece even during the initial high-gravity portion of the parabola. Upon entering the low-gravity portion of the parabola ($t > 5$ s), the operator set the rotation rate at a specified value. As the aircraft began the pullout from the parabola, the gravitational intensity rose (starting at approximately $t = 25$ s). When the gravitational intensity reached a certain limiting value, the bubble became unstable and broke apart. Thus for a fixed rotation rate, there was an upper limit to the gravitational intensity for which the bubble remained stable.

Once an acceleration had been applied to the system that was greater than the limiting gravitational intensity, the system actually took a finite amount of time to break apart. Acceleration data were available at a frequency of 3 Hz from the voltmeter monitoring the accelerometer and we believe that the response time of the system was faster than this rate. For this reason, the limiting gravitational intensity was taken as the average of the acceleration level at breakup and the preceding acceleration level ($\frac{1}{3}$ s earlier) with an error assigned of half the interval.

If the response of the system was actually slower than $\frac{1}{3}$ s (the data acquisition time) then the gravitational intensities measured by the above procedure would be too large since the gravitational intensity was increasing during the measurement. However, the results of the variable-gravity experiments agree with the results of the ground-based studies, where the rotation rate was decreased so slowly that the limiting rotation rate could be determined accurately without worrying about the time response problem.

The breakup of the vapour phase was studied using the above two methods for three different liquids: octyl alcohol, n-hexadecane and water.

3.4. *Experimental observation of the position and orientation of the bubble*

In order to observe the position and orientation of the bubble, a larger cylinder with a diameter of 70 mm and a length of 140 mm was used. The cylinder was partially filled with water, grasped at one end and rotated. The other end was made of glass so that the bubble could be viewed from the end. At low rotation rates the bubble moved off the axis of rotation and it was clear that the cross-section was no longer circular. The bubble formed a rounded 'airfoil-type' shape in which the trailing edge was sharper than the leading edge. Surface tension does not allow the bubble cross-section to assume a true airfoil shape with a sharp trailing edge so that the shape of the cross-section can also be described as an ellipse that is broadened at the leading end.

The position of the bubble was observed at three different rotation rates. A line was drawn from the approximate centre of the bubble cross-section to the centre of the rotating cylinder. The angle that this line makes with the horizontal will correspond to the angle α in figure 1. For the rotating water system, α was measured at rotation rates of 9.6, 14.0 and 19.9 Hz and was found to be $44 \pm 1°$.

At the two lower rotation rates, the orientation of the airfoil shape in the flow was measured as the angle the chord of the airfoil makes with the horizontal. If we assume that the bubble has oriented itself along the direction of flow and thus in the direction of the viscous shear, this angle will correspond to the angle β.

3.5. *Results: comparison of the measured and predicted lengths of the phase boundary*

The predicted lengths of the bubble in the absence of gravity for each of the experimental conditions considered are shown in figure 7 as solid lines. Of the four approximations that were made ((10), (11) and (16) and neglecting gravitational effects), only the one that neglected gravitational effects is of questionable validity in the experiments. For all of the experimental data shown in figure 7, the parameter Γ in (29) is small compared to unity. The maximum value that this parameter reaches for all experimental values used is 0.06 which occurs at one point along the phase boundary at the lowest rotation rate, 4.2 Hz. More typically, for most locations on the

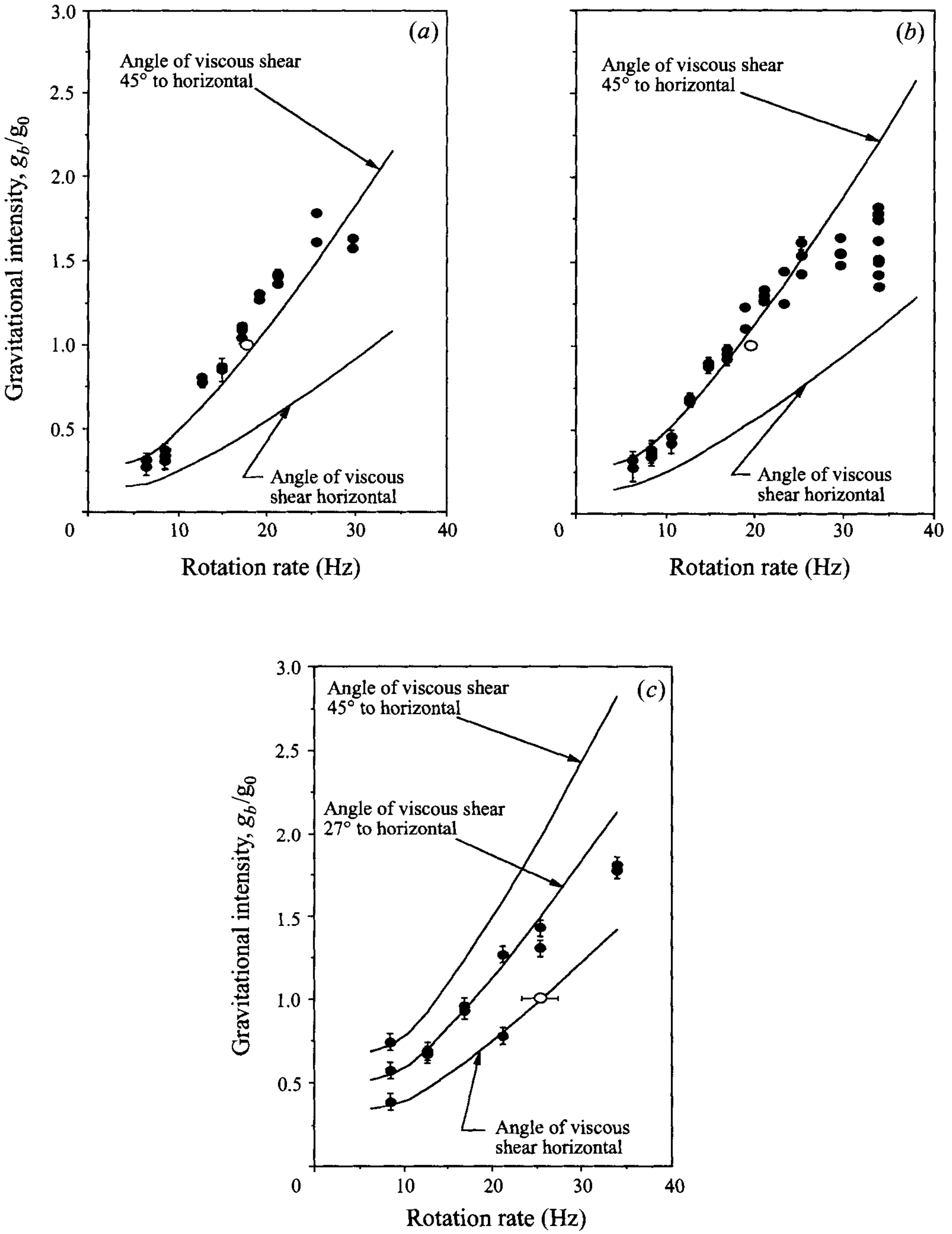


FIGURE 9. Comparison of measured gravitational intensity at breakup (data points) with theoretical gravitational intensity (solid lines) at which zero mean curvature is expected. Comparison is for (*a*) 37.6 g of octyl alcohol in a volume of 46.82 ml, (*b*) 34.7 g of n-hexadecane in a volume of 45.89 ml, (*c*) 44.85 g of water in a volume of 45.89 ml. The open circle represents data obtained on the ground.

bubble surface and at higher rotation rates, this parameter may be on the order of 0.0001 or even lower. The experimental measurements are shown as the data points in figure 7.

In developing the equations for the shape of the phase boundary we assumed that the effect of gravity was negligible. We then used the derived equations along with the experimental conditions to evaluate the parameter Γ and found that it was appropriately small. Since our equations are then consistent with the experimental conditions, we would expect the equations to be able to predict the observed lengths of the vapour phase and indeed, as can be seen in figure 7, the agreement between prediction and measurement is very good.

3.6. *Results: comparison of the measured and predicted breakup of the vapour phase*

In figures 9(*a*)–9(*c*), experimental data for the breakup of three different liquid–vapour systems are compared with the predictions made from (52). The first two liquids, octyl alcohol and n-hexadecane, have relatively low surface tensions of approximately 0.027 N m^{-1}. We can see by looking at figures 9(*a*) and 9(*b*) that the approximation that the surface is free to change its shape and orient itself in the flow field (so that $\beta \approx 45°$) appears to be reasonable for rotation rates below about 25 Hz. At larger rotation rates, the radius of the bubble becomes smaller, increasing the effects of surface tension. Thus the bubble will not be as free to change shape at these higher rotation rates. This has the effect of lowering the angle β.

In figure 9(*c*), water data from an earlier experiment (Yee *et al.* 1991) is compared with the predictions. Water has a higher surface tension (0.072 N m^{-1}) than the previous two liquids. Thus we would expect that the angle of the viscous shear force will be less than 45°. A rotating cylinder containing water was viewed from the end, and the angle that the chord of the airfoil shape made with the horizontal was measured (see §3.4). This angle was 27° when the water was rotating at two different speeds of 9.3 and 14.0 Hz. In figure 9(*c*), the predictions are shown for the three angles of 45°, 27° and 0°. The prediction using the measured angle of $\beta = 27°$ agrees well with the experiments.

4. Discussion and conclusions

In a system such as that considered here, the solution of the Laplace equation, (30), is often referred to as the 'exact' solution for the phase boundary shape (e.g. see Cayias *et al.* 1975; Princen *et al.* 1967; Torza 1975). Such a solution is consistent with the ideas of continuum mechanics. The continuum mechanics concept requires only that the equilibrium pressure be such that a force balance exists at the phase boundary. However, the thermodynamic concept of pressure requires that the phase boundary shape be such that both a force balance exists and there is no net mass flux at the boundary. This latter condition is imposed by requiring the equality of the chemical potentials in the respective phases (equation (6)); however, for an isothermal system, the chemical potential of a bulk phase may be expressed in terms of the pressure that exists in that phase. Thus, the equality of the chemical potentials imposes a second relation on the pressures in the respective phases.

As indicated in §2.1.1, taking into account the equilibrium condition imposed by the equality of the chemical potentials has a significant advantage, in that it allows the equilibrium phase boundary shape to be predicted in terms of the experimentally controllable variables. In fact, the method leads to the possibility of two equilibrium shapes existing in the system; however, this result appears to be physically correct. The smaller size is thermodynamically unstable and represents the threshold size that must be produced in the nucleation of the vapour phase if the system is initially a homogeneous liquid phase. The larger size represents a thermodynamically stable state and is the size to which the vapour phase would grow following the nucleation of this phase from a homogeneous liquid phase (Forest & Ward 1977, 1978; Tucker & Ward 1975; Ward *et al.* 1970).

As the necessary condition for neglecting gravitational effects, it has been proposed that the parameter Γ in (29) must be small compared with unity. This criterion has the significant advantage that it may be evaluated from experimentally controllable variables. Experiments were performed in a reduced gravity environment where the parameter Γ was indeed small. The predictions of the lengths of the vapour phase agreed very well with the measurements made in this environment.

From the point of view of inferring the surface tension from a measured phase boundary shape, the method of solution proposed herein has an advantage, as well, because this solution only contains the surface tension as a parameter. Thus, if the procedure were inverted and the surface tension were inferred by using it as a fitting parameter, there would only be one fitting parameter in the procedure. By contrast, in the previous techniques for inferring the surface tension, the condition of the chemical potentials in the respective phases being equal under equilibrium conditions has not been imposed (Cayias *et al.* 1975; Currie & Van Nieuwkoop 1982; Princen *et al.* 1967; Rosenthal 1962; Vonnegut 1942). To replace this condition for equilibrium, an empirical parameter was introduced. For example, Rosenthal (1962) parameterized the problem so the surface tension could be inferred from knowledge of the phase boundary profile and the measured bubble width; Princen *et al.* (1967) obtained an explicit expression for the surface tension in terms of the measured length of the lighter phase and its volume. Cayias *et al.* (1975) obtained a similar solution, but parameterized the problem so that the surface tension was expressed in terms of the length and width of the lighter phase. As has been seen above, when all of the conditions for equilibrium are imposed, only one empirical parameter can be assigned. Thus, one of the empirical parameters assigned in previous studies is, in fact, determined by the values of the experimentally controllable variables and is not available for assignment.

A theoretical condition for the breakup of the vapour phase in a rotating two-phase system subjected to gravity has been developed. This condition relies on the previously developed theory for the equilibrium shape of a rotating two-phase system in the absence of gravity. The breakup condition was found from the requirement that there exist a point of zero mean curvature somewhere on the interphase. Since the condition is evaluated using calculated values from the axisymmetric theory, the condition is in terms of experimentally controllable parameters and the direction of action of the viscous shear. By making assumptions about the direction of the viscous shear, the theory can be compared with experimental results. In the limit of a long cylinder with low surface tension the thermodynamic approach yields the same stability condition as that found earlier from fluid mechanics (Gans 1977).

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