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Breakage of a drop of inviscid fluid due to a pressure fluctuation at its surface

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In this work, an attempt is made to gain a better understanding of the breakage of low-viscosity drops in turbulent flows by determining the dynamics of deformation of an inviscid drop in response to a pressure variation acting on the drop surface. Known scaling relationships between wavenumbers and frequencies, and between pressure fluctuations and velocity fluctuations in the inertial subrange are used in characterizing the pressure fluctuation. The existence of a maximum stable drop diameter d_{max} follows once scaling laws of turbulent flow are used to correlate the magnitude of the disruptive forces with the duration for which they act.

Two undetermined dimensionless quantities, both of order unity, appear in the equations of continuity, motion, and the boundary conditions in terms of pressure fluctuations applied on the surface. One is a constant of proportionality relating root-mean-square values of pressure and velocity differences between two points separated by a distance l. The other is a Weber number based on turbulent stresses acting on the drop and the resisting stresses in the drop due to interfacial tension. The former is set equal to 1, and the latter is determined by studying the interaction of a drop of diameter equal to d_{max} with a pressure fluctuation of length scale equal to the drop diameter. The model is then used to study the breakage of drops of diameter greater than d_{max} and those with densities different from that of the suspending fluid.

It is found that, at least during breakage of a drop of diameter greater than d_{max} by interaction with a fluctuation of equal length scale, a satellite drop is always formed between two larger drops. When very large drops are broken by smaller-length-scale fluctuations, highly deformed shapes are produced suggesting the possibility of further fragmentation due to instabilities. The model predicts that as the dispersed-phase density increases, d_{max} decreases.

1. Introduction

The breakage of liquid drops suspended in another liquid phase has been studied by many workers since the pioneering work of Taylor (1934). Taylor studied the behaviour of drops in well-defined flow fields at low Reynolds numbers both experimentally and theoretically. Drop behaviour in low-Reynolds-number flows has since been studied extensively using more sophisticated experiments (Bentley & Leal 1986) as well as exact numerical computations (Stone, Bentley & Leal 1986; Stone & Leal 1989). The breakage of drops in turbulent flows is also of considerable interest since turbulent liquid–liquid dispersions are used extensively in the chemical industry for effecting mass and/or heat transfer between two immiscible liquids. In contrast to

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drop behaviour in low-Reynolds-number flows, though considerable work has been done to investigate drop breakup in turbulent flows, the understanding of the process is still at an elementary stage.

The earliest known theoretical treatment of drop breakup in turbulent flows has been for the case of low-viscosity dispersed phase. Kolmogorov (1949, cited by Levich 1962) and Hinze (1955) argued that drops larger than a certain size d_{max} cannot exist indefinitely in a turbulent flow. They proposed that a drop of size l breaks when the pressure fluctuations across the drop caused by velocity fluctuations exceed the forces due to surface tension that resist breakage, i.e.

$$\rho_c \overline{u^2(l)} l^2 > \sigma l, \tag{1.1}$$

where $\overline{u^2(l)}$ is the mean-square relative velocity between two points separated by a distance l, and σ is the interfacial tension between the two phases. Here, the same length scale is used for the size of the eddy and the drop since it is assumed that a drop can be broken most easily by an eddy of size equal to its diameter. This of course is consistent with the generally accepted principle that eddies transfer most of their energy to eddies that are nearly of their own size. Batchelor (1967) has given an expression for $\overline{u^2(l)}$ valid in the inertial subrange as

$$\overline{u^2(l)} \sim (\epsilon l)^{2/3}, \tag{1.2}$$

where ϵ is the power dissipation per unit mass. Substituting this, one arrives at the estimate for d_{max}:

$$d_{max} \sim \frac{(\sigma/\rho_c)^{3/5}}{\epsilon^{2/5}}. \tag{1.3}$$

The above expression has been widely used, and the proportionality constant has been experimentally determined for stirred vessels and other important equipment.

While this approach successfully predicts d_{max}, it does not throw any light on the process of breakage itself. Thus, it is not clear whether the size of the drop and the length scale of velocity fluctuation l should be equal for breakage to occur. Further, neither the time required for fragmentation of a drop bigger in size than d_{max} nor the size distribution of daughter droplets formed by breakage is known. The number and sizes of the daughter droplets formed depend on the course taken by the drop as it extracts energy from the surroundings on its way to breakage and are not obtained within the framework of the existing models. Calculation of breakage frequency involves determination of time of breakage as well as the frequency with which a drop encounters eddies that will succeed in breaking it. It has often been assumed that breakage results in two daughter droplets of equal size (Laso, Steiner & Hartland 1987). If this were the case, one would expect the drop size distribution to have one peak corresponding to the daughter droplets formed by the breakage of drops of diameter greater than d_{max}. In the absence of formation of satellite droplets of any other size, there would be no drops of volume smaller than $v_{max}/2$ where v_{max} is the volume of a drop of diameter d_{max}. These expectations are not supported by experimental observations. Further, there is experimental evidence in favour of formation of multiple daughter droplets in a breakage event. Hinze (1955) has shown that at high Weber numbers drops disintegrate in a rather chaotic manner. His experiments show a large number of daughter droplets being formed.

Information regarding both breakage frequency and daughter droplet size distribution is necessary to determine the drop size distribution. Mechanistic models do exist for dispersed phases of moderate and high viscosity for calculating the breakage frequency. They are one-dimensional in nature and contain *ad hoc* assumptions

regarding breakage criteria and mode of breakage (Arai *et al.* 1977; Lagisetty *et al.* 1986; Nambiar *et al.* 1992). Thus, there is a need to address these issues from a more fundamental viewpoint.

The essential feature of the work of Kolmogorov and Hinze is the visualization of drop breakage in turbulent flows as a result of deformation of drops forced by turbulent pressure fluctuations. In a turbulent flow field, if a pressure fluctuation on the surface of the drop acts for the lifetime of an eddy that deforms it in some direction, it is unlikely that the next pressure fluctuation on the surface of the drop will act in a coherent manner to continue the deformation process. Drop–eddy interactions occur in a random manner and not in any cooperative fashion. It seems, therefore, likely that a drop breaks under the influence of a single fluctuation. The fact that the successful theoretical estimates provided by Kolmogorov and Hinze for inviscid drops do not involve any time scale (except perhaps that implied by ϵ, and l) also seems to suggest the same. In modelling the breakage of viscous drops, Lagisetty *et al.* (1986) and Davies (1985) have also used the lifetime of an eddy as the scaling parameter for estimating the effect of viscosity of the dispersed phase. The success of these models also indicates that even moderately viscous drops perhaps break under the influence of a single pressure fluctuation. Given the complexity of turbulent flows, therefore, one possible approach to understand more about drop breakage in turbulence is to study the deformation of drops under the influence of a single external pressure fluctuation. This viewpoint is explored in the present work.

In this work, an attempt has been made to study the deformation and breakage of an inviscid drop under the influence of a pressure variation that occurs on its surface. While simplifying assumptions have been made for tractability, the evolution of the shape of a drop under the action of external stresses is followed until it breaks into two or more fragments. Thus, an arbitrary breakage condition is not necessary, and it is also possible to learn about the number of daughter droplets formed in a breakage event.

Another issue of fundamental interest is the existence of a maximum stable drop diameter. This has been questioned recently by Lam *et al.* (1996), who found that the value of d_{max} decreases with the duration of the experiment. They argue that, due to intermittencies in turbulence, pressure fluctuations much greater than the root-mean-square value can occur, and given enough time, drops of any size will eventually encounter disruptive forces large enough to break them. However, the very large disruptive forces generated by intermittency are also expected to have a shorter lifespan than the corresponding root-mean-square value. Given that drop fragmentation does require a finite amount of time, it is not clear that intermittency *per se* rules out the existence of d_{max}. We show in the present work that the existence of d_{max} follows once scaling laws of turbulent flow are used to correlate the magnitude of the disruptive forces with the duration for which they act.

2. Drop behaviour under the influence of a pressure field

In this section, we study the behaviour of an inviscid liquid drop under the influence of a pressure variation imposed on its surface for a certain period of time. For this purpose we need to specify the magnitude, shape and duration of the pressure fluctuation as well as the initial state of deformation in the drop. For concreteness, we choose conditions and parametric values typical for turbulent flow.

The sizes of the smallest drops that can be broken by turbulent eddies lie in the inertial subrange (Levich 1962). We therefore consider a pressure fluctuation with a

length scale l belonging to the inertial subrange. The pressure difference across such an eddy of size l is then assumed to be proportional to $\rho_c \overline{u^2(l)}$ where $\overline{u^2(l)}$ is the mean-square relative velocity between two points separated by a distance l. As a simple case, since solving a three-dimensional problem is computationally expensive, we consider only axisymmetric fluctuations. We make a further simplification that the pressure variation on the drop surface is symmetric about its equator. More precisely, the pressure fluctuation on the surface of a drop is represented by

$$p^{ext}(z, t) = c\rho_c \overline{u^2(l)} \sin(\omega t) \cos(2\pi z/l); \quad 0 \leqslant t \leqslant 2\pi/\omega, \tag{2.1}$$

where z is the distance from the equator of the drop, and c is a constant yet to be determined. Batchelor (1967) has given an expression for $\overline{u^2(l)}$ valid in the inertial subrange as

$$\overline{u^2(l)} = 4.82\alpha(\epsilon l)^{2/3}. \tag{2.2}$$

In (2.1), $2\pi/\omega$ is the duration of a pressure fluctuation of wavelength l (wavenumber = $2\pi/l$). As we adopt parameters typical of high-Reynolds-number turbulent flow, we follow the appropriate scaling rules. Accordingly, we identify ω with the frequency, $\omega(\kappa)$ associated with an eddy of wavenumber κ. Tennekes & Lumley (1972) give the relationship,

$$\omega(\kappa) = \alpha^{1/2} \epsilon^{1/3} \kappa^{2/3}, \tag{2.3}$$

where ϵ is the energy dissipation rate per unit mass and α is the constant of proportionality in the expression for the energy spectrum in the inertial subrange, given by the equation

$$E(\kappa) = \alpha \epsilon^{2/3} \kappa^{-5/3}, \tag{2.4}$$

and has a value of approximately 1.5 (Batchelor 1967). In terms of the length scale, the frequency is given by $\alpha^{1/2} \epsilon^{1/3} (2\pi/l)^{2/3}$.

Nothing can be said *a priori* about the initial deformation and the velocity field inside the drop as it encounters successive pressure fluctuations in a turbulent flow. We therefore study the response of an undeformed drop to the action of the pressure fluctuation acting on its surface.

2.1. *Governing equations*

It is assumed that the flow is irrotational inside the drop. The equations of continuity and motion then become

$$\nabla \cdot \boldsymbol{v} = 0, \tag{2.5}$$

$$\frac{\partial \boldsymbol{v}}{\partial t} + \boldsymbol{v} \cdot \nabla \boldsymbol{v} = -\frac{1}{\rho_d} \nabla p, \tag{2.6}$$

where ρ_d is the density of the dispersed phase.

As the flow is assumed to be irrotational inside the drop, the velocity is written as the gradient of a potential, i.e. $\boldsymbol{v} = \nabla\phi$. Substituting this for $\boldsymbol{v}$ in the equations of continuity and motion gives

$$\nabla^2 \phi = 0, \tag{2.7}$$

$$\nabla \frac{\partial \phi}{\partial t} + \tfrac{1}{2} \nabla (\nabla\phi \cdot \nabla\phi) = -\nabla \left(\frac{p}{\rho_d} \right). \tag{2.8}$$

It follows from (2.8) that

$$\frac{\partial \phi}{\partial t} + \tfrac{1}{2} (\nabla\phi \cdot \nabla\phi) + \frac{p}{\rho_d} = f(t). \tag{2.9}$$

As the drop fluid is incompressible, it is sufficient to determine only the spatial gradients of pressure rather than the absolute values. As this is true of ϕ as well, the right-hand side of (2.9) can be set equal to zero. This amounts to determining the velocity potential and pressure within an arbitrary additive constant. Thus,

$$\frac{\partial \phi}{\partial t} + \tfrac{1}{2}\nabla(\nabla\phi \cdot \nabla\phi) + \frac{p}{\rho_d} = 0. \tag{2.10}$$

2.2. *Boundary condition*

As the drop fluid is inviscid and the stress acting on the surface of the drop by the external fluid is $p^{ext}(R(\theta, t), \theta, t)$, the normal stress balance at the drop surface is

$$p(R(\theta, t), \theta, t) = p^{ext}(R(\theta, t), \theta, t) + \sigma \nabla_s \cdot \boldsymbol{n}, \quad 0 \leqslant t \leqslant 2\pi/\omega, \tag{2.11}$$

where $r = R(\theta, t)$ is the shape of the drop surface, $\boldsymbol{n}$ is the unit outward normal on the surface and the surface gradient operator ∇_s is given by

$$\nabla_s = (\boldsymbol{I} - \boldsymbol{nn}) \cdot \nabla.$$

2.3. *Initial conditions*

For reasons that have been explained earlier, the following initial conditions are used:

$$\text{at } t = 0, \quad \phi = 0, \qquad 0 \leqslant r \leqslant a, \quad 0 \leqslant \theta \leqslant \pi \tag{2.12}$$

and

$$R(\theta, 0) = a. \tag{2.13}$$

Given ϕ on the surface of the drop at any given time, (2.7) can in principle be solved to determine ϕ everywhere inside the drop. The velocity field can then be obtained. However, it is possible to compute with considerably less effort the velocities on the surface, using the boundary integral method. Since we are interested in determining the shape of the drop as a function of time, it is sufficient to compute surface velocities. In order to trace the history of the drop, however, it is necessary to know how the boundary condition varies with time, i.e. how ϕ varies with time on the boundary. This is obtained from (2.10) which is valid everywhere in the drop including on its surface. Applying this equation to any point on the surface by substituting for p from the normal stress balance (2.11), we obtain

$$\frac{\partial \phi(R(\theta, t), \theta, t)}{\partial t} + \tfrac{1}{2}\{\nabla\phi(R(\theta, t), \theta, t) \cdot \nabla\phi(R(\theta, t), \theta, t)\} + \frac{p^{ext}(R(\theta, t), \theta, t) + \sigma \nabla_s \cdot \boldsymbol{n}}{\rho_d} = 0. \tag{2.14}$$

This is solved for $\phi(R(\theta, t), \theta, t)$ with the initial conditions

$$\phi(R(\theta, 0), \theta, 0) = 0, \quad R(\theta, 0) = a. \tag{2.15}$$

The evolution of the shape of the drop in time can thus be traced for any given external pressure distribution $p^{ext}(\theta, t)$.

2.4. *Non-dimensionalization of the governing equations*

The length scale l is chosen as the characteristic length and $(\overline{u^2(l)})^{1/2}$, $l/(\overline{u^2(l)})^{1/2}$ and $\rho_c \overline{u^2(l)}$ as the characteristic velocity, time and pressure, respectively. With the same symbols as in the dimensional equation (2.14), the dimensionless equation then is

$$\frac{\partial \phi(R(\theta, t), \theta, t)}{\partial t} + \tfrac{1}{2}\{\nabla\phi(R(\theta, t), \theta, t) \cdot \nabla\phi(R(\theta, t), \theta, t)\} + \frac{\rho_c}{\rho_d}\left\{p^{ext}(R(\theta, t), \theta, t) + \frac{\sigma}{\rho_c l \overline{u^2(l)}} \nabla_s \cdot \boldsymbol{n}\right\} = 0, \tag{2.16}$$

where the dimensionless external pressure and the dimensionless frequency are given by

$$p^{ext}(R(\theta,t),\theta,t) = c\sin(\omega t)\cos(2\pi z) \tag{2.17}$$

and

$$\omega = \frac{\alpha^{1/2}\epsilon^{1/3}(2\pi/l)^{2/3}l}{(\overline{u^2(l)})^{1/2}} = \frac{(2\pi)^{2/3}}{\sqrt{4.82}}. \tag{2.18}$$

The dimensionless initial condition is

$$\phi(R(\theta,0),\theta,0) = 0, \quad R(\theta,0) = \tfrac{1}{2}d/l, \tag{2.19}$$

where d is the diameter of the drop.

Equation (2.16) has three dimensionless parameters: the constant of proportionality c which is contained in p^{ext} (see 2.1)), the group $\rho_c l\overline{u^2(l)}/\sigma$ which is a Weber number, and the density ratio ρ_c/ρ_d. In addition, the initial condition has a dimensionless parameter, namely the ratio d/l. Using (2.2), we obtain for the Weber number, the equation

$$We = \frac{4.82\alpha\rho_c\epsilon^{2/3}l^{5/3}}{\sigma}. \tag{2.20}$$

The local energy dissipation rate per unit mass, ϵ, depends on the geometric and kinematic variables pertaining to the turbulent flow of interest. It is now necessary to explicitly bring into the equations the diameter of the drop, d. Thus, we express We as

$$We = \frac{4.82\alpha\rho_c\epsilon^{2/3}d^{5/3}(l/d)^{5/3}}{\sigma}. \tag{2.21}$$

Substituting for the Weber number in (2.16), we obtain

$$\frac{\partial\phi(R(\theta,t),\theta,t)}{\partial t} + \tfrac{1}{2}[\nabla\phi(R(\theta,t),\theta,t)\cdot\nabla\phi(R(\theta,t),\theta,t)]$$
$$+\frac{\rho_c}{\rho_d}\left\{p^{ext}(R(\theta,t),\theta,t) + \frac{1}{4.82\alpha}\frac{\sigma}{\rho_c\epsilon^{2/3}d^{5/3}}\left(\frac{d}{l}\right)^{5/3}\nabla_s\cdot\boldsymbol{n}\right\} = 0. \tag{2.22}$$

The dimensionless angular frequency ω is determined by (2.18). This gives a value of approximately 0.247 for the dimensionless frequency $\omega/2\pi$. We fix this value at $\frac{1}{4}$. The angular frequency ω is therefore equal to $2\pi/4$. There are two other undetermined dimensionless quantities in (2.22). These are the dimensionless amplitude of the pressure fluctuations c, which is contained in p^{ext}, and the Weber number We defined by (2.20). The considerations based on which the values of these quantities are fixed are explained below.

2.5. *The concept of the maximum stable drop diameter*

Equation (2.22) has two undetermined constants, namely c, which is contained in p^{ext}, and ϵ, which is contained in We defined by (2.20). Both these constants appear in the terms enclosed by curly braces on the left-hand side of (2.22). During the first quarter-cycle of the imposed fluctuation, the gradient of the external pressure tends to elongate and break the drop. In general, till the drop gets pinched off somewhere, surface tension tends to restore the spherical shape. The first term inside the braces of (2.22) represents the forces tending to deform the drop while the second term represents the forces resisting the former. The magnitudes of the two terms are determined by the constants c and We. It can therefore be concluded from (2.22) that increasing c or We leads to larger deformation and hence to easier breakage. Thus, for a given value of c and d/l, there exists a minimum value of We which leads to breakage of the drop.

The validity of this argument is of course contingent upon the validity of the scaling

relationships among the various quantities that have been used in non-dimensionalizing the governing equations. These relationships are expected to apply to high-Reynolds-number turbulent flows. As stated earlier, the concept of maximum stable drop diameter in turbulent flows is linked to the condition where the drop diameter and length scale of the pressure fluctuation are equal. By setting $d = l$, the concept of maximum stable drop diameter readily follows from the above argument. Since $We = \sigma/(\rho_c \epsilon^{2/3} d^{5/3})$, there exists a minimum value for the diameter of the drops which can be broken, and the value can be determined once the two constants c and ϵ are known.

We can now examine the applicability of the above arguments in the presence of large pressure fluctuations caused by intermittency. In the above arguments, we have used ϵ, $\overline{u^2(l)}$, and ω, which represent temporal averages of these quantities. Actually, each of these quantities has a distribution of values, i.e. an eddy of a given size has a distribution of energies and frequencies. These quantities are correlated as $\overline{u^2(l)} \sim (\epsilon l)^{2/3}$ and $\omega \sim \epsilon^{1/3} l^{-2/3}$. This is equivalent to $\omega \sim (\overline{u^2(l)})^{1/2}/l$. Let us assume that the constants of proportionality in the correlations between $\overline{u^2(l)}$ and the pressure fluctuation, as well as in the correlations of ϵ with $\overline{u^2(l)}$ and ω remain valid for any single period of the fluctuation. Then, the scaling rule $\omega \sim (\overline{u^2(l)})^{1/2}/l$ remains valid for any single fluctuation as well, and the dimensionless frequency will always have the same value. Now if instead of $(\overline{u^2(l)})^{1/2}$ a value of velocity fluctuation characteristic of dissipation in a single period of fluctuation is used for the purposes of non-dimensionalization, the dimensionless equations also remain unaltered. Thus, the minimum value of We established also remains unaltered. However, with the new scaling, ϵ in $We = \sigma/(\rho_c \epsilon^{2/3} d^{5/3})$ is the dissipation rate characteristic of a single fluctuation. Hence, as long as ϵ remains bounded, a value for the maximum stable drop diameter will exist, and it will be lower than that calculated using $\overline{u^2(l)}$. Moreover, as the frequency of occurrence of fluctuations in power dissipation decreases as the magnitude of the fluctuation increases, it is quite likely that the observed value will decrease slowly with the duration of the experiment.

If we call the maximum stable drop diameter d_{max} and the Weber number corresponding to a pressure fluctuation of length scale equal to d_{max} We_{min}, we can write

$$We_{min} = 4.82\alpha\rho_c \epsilon^{2/3} d_{max}^{5/3}/\sigma. \tag{2.23}$$

In the present case, the constants c and We_{min} are both unknown. However, if either of them is fixed arbitrarily, the other can be determined by doing computations for interactions between a drop of diameter d_{max} and a pressure fluctuation of length scale d_{max}. Alternatively, if We is fixed, the minimum value of c for which breakage occurs can be determined. The former alternative was chosen in the present case. The order of magnitude of c can be obtained from the following relationship derived by Oboukhov (1949, quoted in Batchelor 1967, p. 183):

$$\overline{p^2(l)} = \rho^2\{\overline{u_1^2(l)}\}^2. \tag{2.24}$$

Here, $\overline{u_1^2(l)}$ is the mean-square value of the longitudinal component of the relative velocity between two points separated by a distance l. This suggests that the value of c must also be of order unity. Hence, we set $c = 1$ in our model.

Once we associate We_{min} with a pressure fluctuation of length scale d_{max}, the Weber number corresponding to any such pressure fluctuation in the inertial subrange can be calculated. As can be seen from (2.20), the Weber number corresponding to a pressure fluctuation of length scale x times d_{max} is $x^{5/3}$ We_{min}. We use this relationship to study the behaviour of drops of size other than d_{max}.

3. Solution of the governing equations

As has already been stated, we intend to study the evolution of the shape of a drop as a function of time in response to stresses imposed on the boundary. For this, it is sufficient to determine the velocity of the fluid everywhere on the surface of the drop at any instant. The boundary integral method was therefore chosen for solving the equation for the velocity potential inside the drop. The generalized vortex method developed by Baker, Meiron & Orszag (1980, 1982, 1984) and applied by Lundgren & Mansour (1988) to the study of free oscillations of inviscid drops was used. In this method, the velocity potential is expressed as a function of a surface distribution of dipoles, the density of which can be determined knowing the value of the potential on the surface. The surface distribution of dipoles is equivalent to a vortex sheet as there is a jump in the tangential component of the velocity across the surface. Thus,

$$\phi_P = \int_S \mu \frac{\partial g(\boldsymbol{r}_P, \boldsymbol{r})}{\partial n} \mathrm{d}S, \quad P \notin S, \tag{3.1}$$

where $g(\boldsymbol{r}_P, \boldsymbol{r}) = -1/(4\pi|\boldsymbol{r}_P - \boldsymbol{r}|)$. Here, $\boldsymbol{r}$ represents the position vector of a point, μ the dipole density, suffix P denotes a fixed point P, all quantities without the suffix P refer to any point Q on the boundary S and $\partial/\partial n$ is the derivative in the direction of the outward normal on the boundary at Q. The integration is with respect to Q. Thus, $\boldsymbol{r}$ means $\boldsymbol{r}_Q$, and ϕ_P and μ mean $\phi(\boldsymbol{r}_P)$ and $\mu(\boldsymbol{r}_Q)$, respectively. The procedure described by Lundgren & Mansour (1988) for an axisymmetric problem was used except that ϕ and μ were approximated by piecewise cubic polynomials in arclength s over the boundary. The integrations with respect to time were done using the Runge–Kutta–Gill method.

The length of the time step required for a given order of accuracy decreases as the interface velocity increases and as the spacing between the nodes on the interface decreases. The following empirical equation was used, after every step, to calculate the length Δt of the next time step:

$$\Delta t = \frac{(\Delta t)_{max}}{\left(1.0 + \dfrac{v_{max}}{s_n/n}\right)},$$

where v_{max} is the maximum magnitude of the velocity obtained on the interface from computations for the current time step, s_n is the arclength along any meridian, n is the number of elements into which the meridian is divided and $(\Delta t)_{max}$ is a value Δt will never exceed. By using different values for $(\Delta t)_{max}$, and comparing the results, it was found that a value of 0.005 was adequate and was used in all the computations.

4. Results and discussion

We first present computations made to validate the computer program. We then proceed to present the results of calculations made to fix We_{min}. Finally, we present the effects of various parameters on drop behaviour.

4.1. *Validation of the computer program*

Our program was used to solve the problem of free oscillations of an inviscid drop with a given initial distribution of the velocity potential ϕ on its surface. This problem has been solved by Lundgren & Mansour (1988). Computations were made for one and a quarter periods of oscillation of the drop and our results agreed very well with the

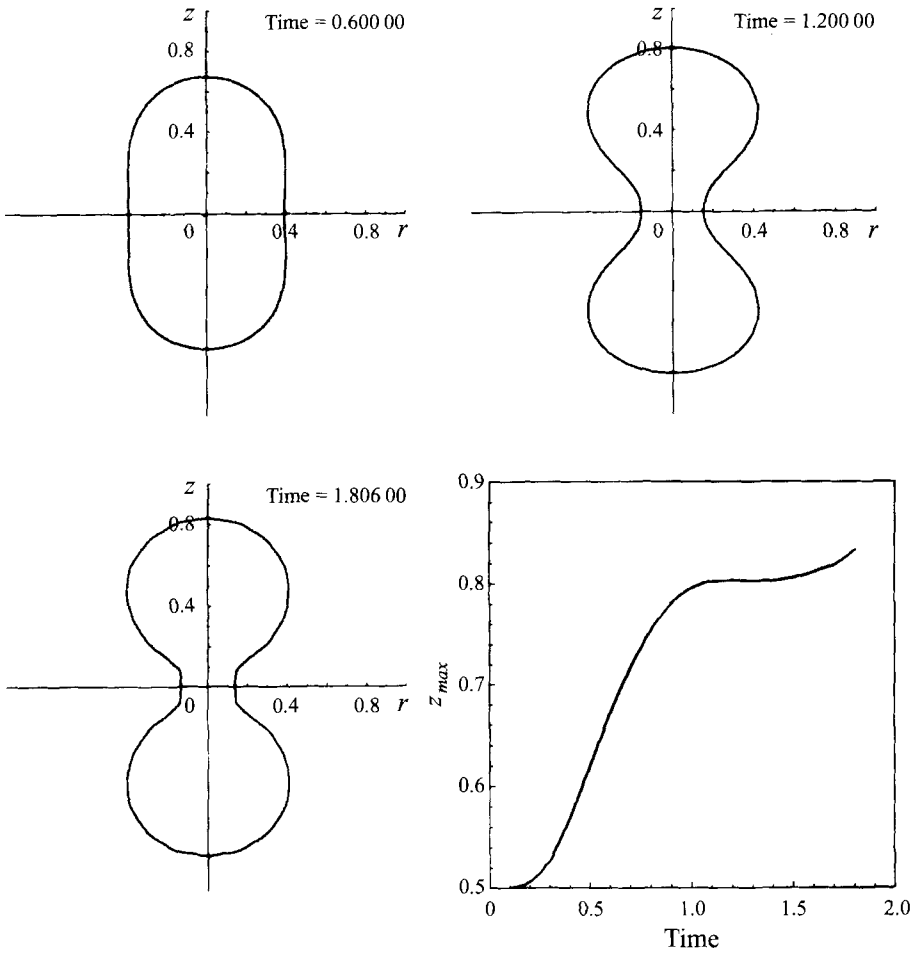

FIGURE 1. The shape of the drop at different times and variation of the distance of the tip of the drop from the equator with time for $c = 1.0$, $\omega = 2\pi/4$, $a/l = 0.5$, $d/d_{max} = 1.0$, $\rho_c/\rho_d = 1.0$ and $We = 2.41$.

graphs presented by Lundgren & Mansour. The computations in the present work were made with piecewise cubic approximations for the unknowns on the boundary, whereas Lundgren & Mansour (1988) approximated the value over each element by that at the midpoint of the element. They used 100 nodes on the boundary. When there is symmetry about the equator, therefore, there are 50 nodes on each half. We used 15 elements on each half of the boundary. The number of numerical integrations needed to assemble the matrix with 50 elements using the method of Lundgren & Mansour (1988) is $50 \times 50 = 2500$, whereas with 15 cubic elements, it is $15 \times 15 \times 4 = 900$. Thus, there is a saving on computational effort. Lundgren & Mansour (1988) used time-smoothing in their work to remove short-wavelength fluctuations. This was not necessary in our computations up to the point at which computations are presented.

4.2. *Determination of* We_{min}

The results of computations made to determine the value of We_{min} are shown in figures 1 and 2. In these figures, the shapes of the drops at different times are shown. The coordinate of the tip of the drop is also shown as a function of time. These

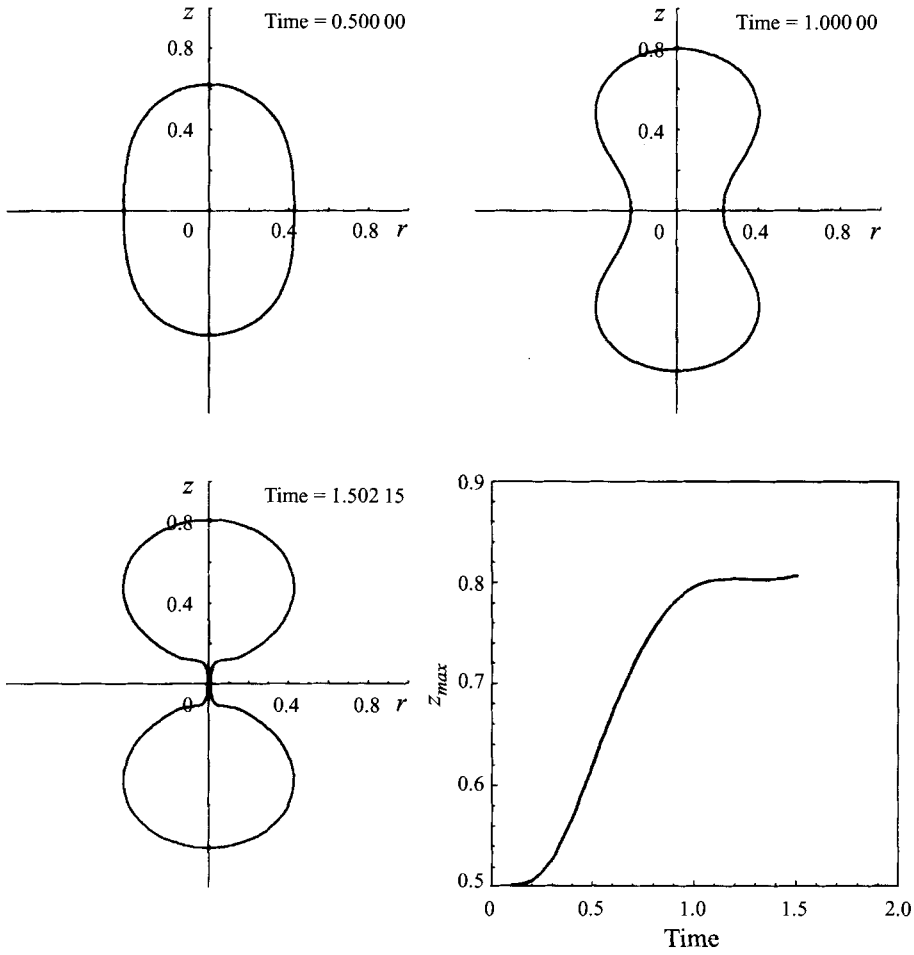

FIGURE 2. As figure 1 but for $We = 2.42$.

computations were made with $c = 1.0$, $\omega = 2\pi/4$ and $\rho_c/\rho_d = 1.0$. As mentioned earlier, the size of the drop and length scale of the pressure variation should be equal for determining d_{max}. Hence, a/l, where a is the radius of the drop, is set equal to $\frac{1}{2}$. As has been stated in §3, at each instant, attention is focused on a material point and integration with respect to time is done by the Runge–Kutta–Gill method. The new position of each material point is therefore obtained from the computed velocity. Just before breakage occurs, the drop forms a neck. At some point in time, the diameter of this neck will be extremely small. At this juncture, long-range and short-range intermolecular forces may become important, and the continuum model itself may become invalid. These details have not been taken into account in the present work. Instead, as numerical integration is done with respect to time, when the interface crosses the axis of symmetry at any point, the drop is assumed to be broken. To some extent, this breakage condition depends on the length of the time steps used. However, to make the computations as objective as possible, the same strategy, described in §3, was used to choose the length of the time steps in all computations.

As the angular frequency ω was chosen to be $2\pi/4$, the time taken for one full cycle of the imposed pressure fluctuation is 4. It is seen from figure 2 that breakage occurs for $We = 2.42$ in the second quarter of this cycle. In contrast with this, for $We = 2.41$,

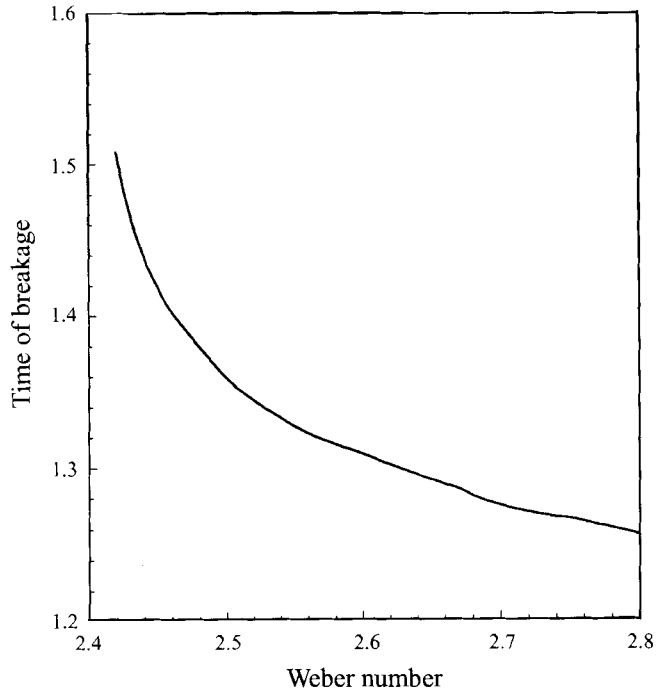

FIGURE 3. Time of breakage as a function of Weber number for $c = 1.0$, $\omega = 2\pi/4$, $a/l = 0.5$, $d/d_{max} = 1.0$ and $\rho_c/\rho_d = 1.0$.

the results of which are presented in figure 1, the diameter of the neck at the equator of the drop reaches a minimum value and then increases. For the form of the fluctuation assumed, during the first half-period the pressure at the equator of the drop is greater than at the neighbouring points on either side. During the second half of the cycle, the situation is reversed. The magnitude of the pressure gradient at the equator of the drop increases during the first quarter of a cycle, reaches a maximum and decreases during the second quarter. Thus, while the external pressure tends to squeeze the drop at the equator during the first half-period, the strength of this squeezing force reaches a maximum at the end of the first quarter and decreases to zero at the end of the second quarter. As the drop is squeezed at the equator, it gets elongated. Consequently, the resistance to deformation due to interfacial tension increases. During the second quarter of the cycle, a point is reached when the resisting force becomes stronger than the squeezing force due to the external pressure. In the absence of inertia, the direction of net flow within the drop would reverse at this instant. This reversal is delayed by the inertia of the drop fluid. Thus, the drop attains a maximum deformation a short time after one quarter of the period of the fluctuation. If the drop does not pinch off at this point, it does not break within the period of the imposed fluctuation. It is seen that the minimum value of We for breakage to occur in the first period is approximately 2.42. We accept this as the minimum value of We required for breakage to occur and call this We_{min}. The value of We_{min} is thus determined within an error of 0.01.

Computations were made with several values of We to arrive at the above value of We_{min}. The time taken for breakage decreases as the Weber number increases as shown in figure 3, as expected. The time of breakage increases more steeply with decrease in We as We_{min} is approached. For $We < We_{min}$, breakage does not occur within one

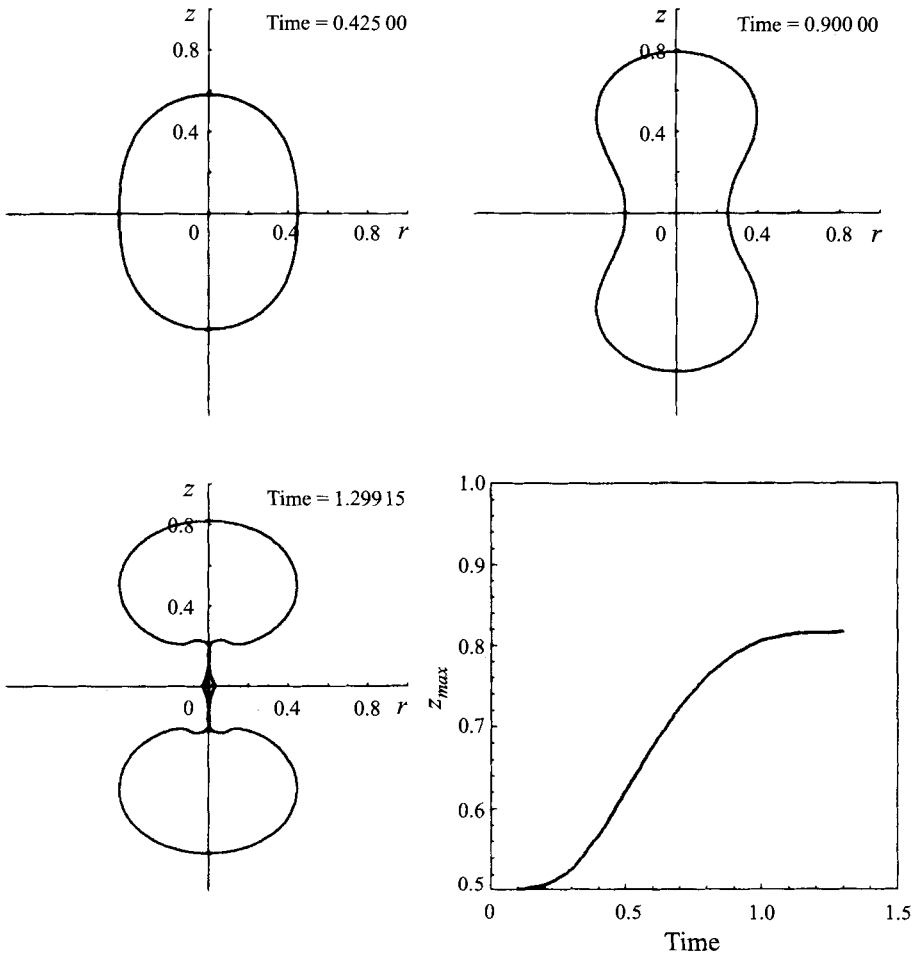

FIGURE 4. The shape of the drop at different times and variation of the distance of the tip of the drop from the equator with time for $c = 1.0$, $\omega = 2\pi/4$, $a/l = 0.5$, $d/d_{max} = 1.050$, $\rho_c/\rho_d = 1.0$ and $We_{min} = 2.42$.

cycle of the pressure fluctuation. As has already been explained, we then consider the drop to be stable. This also admits the following interpretation. Each computation with $We > 2.42$ can be regarded as one with $d/d_{max} > 1$ and $d = l$. For such cases, the time of breakage is smaller and it increases steeply as the drop diameter approaches a value equal to d_{max}. For $d/d_{max} < 1$, the drop does not break within one cycle of the imposed pressure fluctuation. These drops are deemed to be stable under the conditions, i.e. for the prevailing value of ϵ.

4.3. *Breakage of drops of diameter greater than d_{max} with $d = l$*

Accepting 2.42 as the value of We_{min}, we proceed to study the breakage of a drop of diameter greater than d_{max} with $d = l$. The results are shown for $d/d_{max} = 1.05$ in figure 4. It is seen that the dimensionless time of breakage decreases as the drop size is increased beyond d_{max}. Further, breakage occurs farther away from the equator as the drop size increases. Results obtained for $d/d_{max} = 1.025$, though not shown, reveal the same features except that the size of the satellite drop is smaller. It is possible that satellite droplets are always formed during breakage. The formation of satellite

droplets during pinch-off has been reported by Mansour & Lundgren (1990) in their study of inviscid capillary jet breakage and by Tjahjadi, Stone & Ottino (1992) in viscous capillary breakup. Studying satellite drop formation requires very fine resolution of the drop surface near the point of breakage and has not been undertaken in the present work. It is clear, however, that during the breakup of drops of diameter considerably greater than d_{max}, at least one big satellite drop is formed at the centre, the size of which increases as the size of the parent drop increases. This can be interpreted as follows.

Breakage is preceded by the formation of a neck. This is because, as the drop is elongated, the curvature at its ends is greater than at the equator. As the deformation increases, the difference in curvature at these points increases. The higher curvature at the ends drives the fluid towards the equator with a force which increases with time during the phase in which the elongation of the drop increases with time. On the other hand, the squeezing force due to the external stresses first increases, reaches a maximum at the end of a quarter of a cycle and then decreases. When the force due to surface tension gains dominance over the external forces and the inertia of the drop fluid, the drop takes the shape of a dumb-bell, with two bulbous portions connected by a neck. The length of the neck depends on the fraction of the drop in which the restoring force due to interfacial tension dominates over external forces and inertia. Since the amplitude of the pressure fluctuations $\sim d^{2/3}$ and the cross-sectional area of the drop $\sim d^2$, the net forces due to external pressure $\sim d^{8/3}$. The inertia of the drop fluid $\sim d^3$. The force due to interfacial tension $\sim d$, as the curvature $\sim d^{-1}$ and the cross-sectional area $\sim d^2$. Thus, for larger drops, the external forces and inertia are more dominant and, hence, the fraction of the drop in which the forces due to interfacial tension dominates over these forces is smaller. Consequently, the neck is longer and pinching off takes place at a greater distance from the equator.

4.4. *Breakage of drops of diameter greater than d_{max} with $l < d$*

For a drop to break, the energy required to create new surface has to be supplied by the pressure variation on the surface. While surface energy per unit volume $\sim d^{-1}$, the kinetic energy per unit volume generated by the pressure fluctuation $\sim l^{2/3}$. Thus, a drop of diameter greater than d_{max} can be broken by a pressure fluctuation on a smaller length scale. Though Nambiar *et al.* (1992) propose that such a mechanism does exist in turbulent stirred dispersions, there has been no validation of such a hypothesis.

We have investigated the breakage of very large drops by pressure fluctuation on smaller length scales. Figure 5 shows the results of the interaction of a drop of diameter 10 times d_{max} with a fluctuation of wavelength equal to the drop radius, i.e. $a/l = 1$. In this case, the pressure on the drop is compressive at the poles as well as the equator. Therefore, when the drop is being squeezed at the equator, it is also being pushed inward at the poles. The axial length of the drop therefore decreases with time and an umbrella-like shape results, as shown in figure 5. The highly deformed shapes produced suggest the possibility of further breakage taking place due to instabilities which are not axisymmetric. A large number of daughter droplets may then be formed. In this context we might recall the experiments of Hinze (1955) in which breakage results in large number of daughter droplets at high Weber numbers. Drop size distribution data obtained from stirred turbulent dispersions, when fitted to population balance equations, also indicate that large drops break into multiple fragments (Narsimhan, Nejfelt & Ramkrishna 1984). The predictions of the present calculations are in agreement with these observations. Similar results were obtained for a drop of diameter 5 times d_{max}, but they are not shown here.

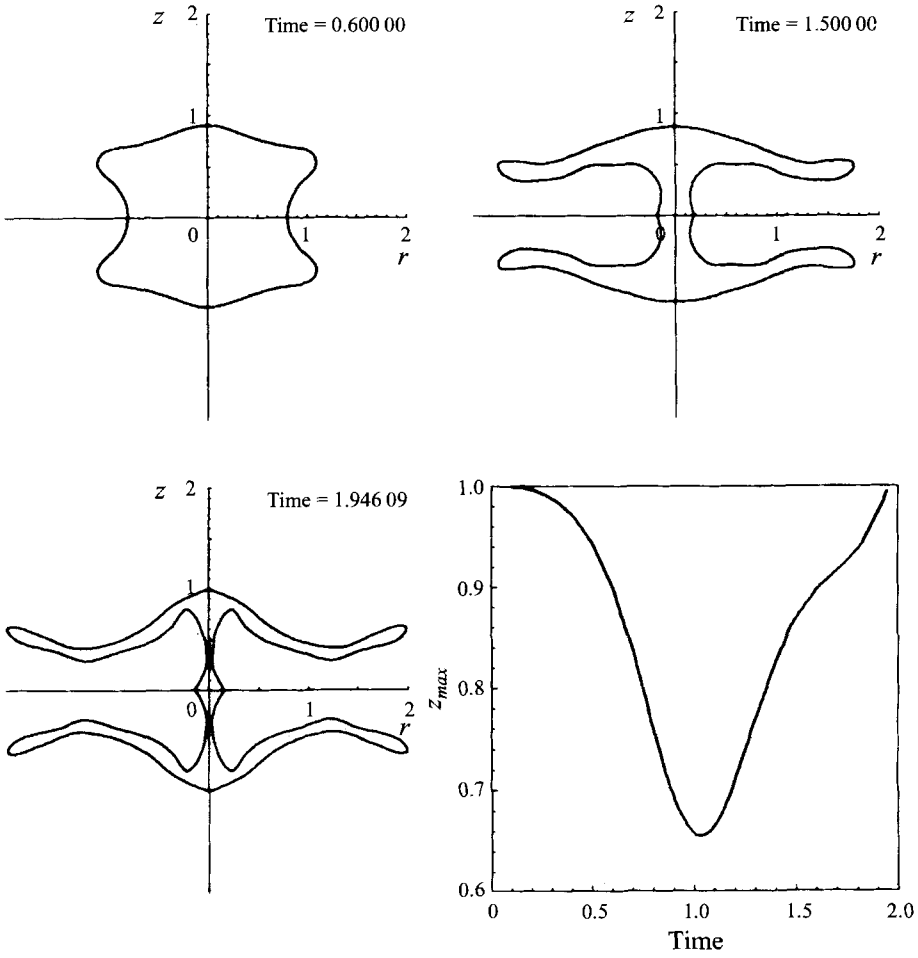

FIGURE 5. The shape of the drop at different times and variation of the distance of the tip of the drop from the equator with time for $c = 1.0$, $\omega = 2\pi/4$, $a/l = 1.0$, $d/d_{max} = 10.0$, $\rho_c/\rho_d = 1.0$ and $We_{min} = 2.42$.

4.5. *Effect of* ρ_c/ρ_d

Another parameter that can affect breakage is the ratio of the densities of the continuous and dispersed phases. Computations were done for cases in which the drop and the suspending fluid have different densities. Here we determine d_{max} by trial and error, assuming that the values of c and We_{min} determined for the case of equal densities are valid.

Computations were done to determine d_{max} for $\rho_c/\rho_d = 0.8$ and 1.2. The deformation patterns of drops were similar to those observed for $\rho_c/\rho_d = 1.0$ and hence are not shown. For the former case, d_{max} was found to lie between 0.98 and 0.99 times $(d_{max})_1$ while for the latter case it was found to lie between 1.01 and 1.02 times $(d_{max})_1$, where $(d_{max})_1$ is the value of d_{max} for $\rho_c/\rho_d = 1.0$. Similar computations were done for $\rho_c/\rho_d = 0.75$ and 1.25. However, since d_{max} can be determined only by trial and error, it was not attempted to resolve it more closely than 0.01 times $(d_{max})_1$. With this resolution, the value of d_{max} for $\rho_c/\rho_d = 0.75$ was found to be in the same interval as for $\rho_c/\rho_d = 0.80$, i.e. between 0.98 and 0.99 times $(d_{max})_1$. Similarly, the value of d_{max}

for $\rho_c/\rho_d = 1.25$ was found to be in the same interval as for $\rho_c/\rho_d = 1.20$, i.e. between 1.01 and 1.02 times $(d_{max})_1$. However, the breakage time was found to increase monotonically with increase in ρ_c/ρ_d, indicating that within this range of the density ratio, d_{max} also increases with increase in ρ_c/ρ_d.

Thus, our calculations indicate that d_{max} decreases with increase in dispersed-phase density. This dependence is weak, and cannot be discriminated experimentally. The effect of density comes through the inertia of the drop fluid and is difficult to predict *a priori*. If the density of the drop fluid is greater than that of the surrounding fluid, its acceleration in the initial stages of its interaction with a pressure fluctuation is lower, but its deceleration in the later stages is also lower. Whether breakage becomes easier or harder depends on which of these two effects is more important. It appears from the result of our computations that the latter is more important. This trend is opposite to what is known to be the case for viscous drops broken in turbulent stirred vessels (Calabrese, Chang & Dang 1986). However, the effect of density difference on d_{max} for the case of low-viscosity drops does not seem to have been reported so far.

The predictions made using this approach of course depend on the form assumed for the pressure fluctuation which affects not only the time required for breakage but also the mode of breakage, i.e. the sizes and shapes of the daughter droplets formed. Thus, while the approach is useful in gaining a better qualitative understanding of the mechanism of breakage, it cannot directly be used for quantitative predictions of drop size distributions.

4.6. *Application to drop breakage in a stirred vessel*

The above model can be applied to the process of breakage of drops in a stirred vessel. This is done using the available correlations for ϵ and $\overline{u^2(l)}$ in a stirred vessel. The local value of ϵ in a stirred vessel depends on the vessel and stirrer dimensions, the stirrer speed and position. For geometrically similar vessels, we have the correlation

$$\epsilon = c' N^3 D^2, \tag{4.1}$$

where N is the impeller speed in rotations per unit time, D is the diameter of the impeller and the constant of proportionality c' is a function of position in the vessel. Substituting for ϵ in the expression for We, we obtain

$$We = 4.82\alpha c' \left(\frac{\rho_c N^2 D^3}{\sigma}\right) \frac{(d/D)^{5/3}}{(d/l)^{5/3}}. \tag{4.2}$$

Setting $d/l = 1$ and $d = d_{max}$, we obtain

$$\frac{d_{max}}{D} = \left(\frac{We_{min}}{4.82\alpha c'}\right)^{3/5} \left(\frac{\sigma}{\rho_c N^2 D^3}\right)^{3/5}, \tag{4.3}$$

which is the well-known relationship first proposed by Hinze (1955) for inviscid drops.

The order of magnitude of We_{min} can be predicted using the following relationship (Lagisetty *et al.* 1986) for d_{max} which is in agreement with experimental data:

$$\frac{d_{max}}{D} = \frac{1}{8}\left(\frac{\sigma}{\rho_c N^2 D^3}\right)^{3/5}, \tag{4.4}$$

Comparing (4.4) with (4.3), we obtain

$$We_{min} = \frac{4.82\alpha c'}{32}. \tag{4.5}$$

Since the energy dissipation rate per unit mass ϵ is a function of position in the stirred vessel, so is c' (see (4.1)). For a vessel fitted with a Rushton turbine impeller, c' has an average value of about 0.4 (Coulaloglou & Tavlarides 1977) and a maximum about 10 times the average. Since, d_{max} and We_{min} are determined by the maximum value of c', using a value of 4 for c' in (4.5) gives We_{min} of order unity, indicating that the forces due to turbulence in the suspending fluid and those due to interfacial tension are of the same order of magnitude. This prediction is confirmed by our computations which yielded a value of 2.42 for We_{min} assuming a value of unity for c.

5. Conclusions

We have studied the deformation and breakage of an inviscid drop of diameter d under the influence of an axisymmetric pressure variation acting on its surface for a fixed period of time. The pressure variation was characterized by a length scale l. We assumed that the magnitude of the pressure variation and its duration correlate according to the scaling observed for length scales lying in the inertial range of turbulent eddies. The equations contain a constant c, which determines the magnitude of the non-dimensional pressure fluctuation, and Weber number We, which represents the ratio of the forces due to surface tension that resist deformation and the pressure forces. For $d = l$ the results show that if c is fixed, there exists a minimum value for Weber number We_{min} below which breakage cannot occur. This implies that for a given level of the pressure fluctuation, drops of size below a certain diameter d_{max} cannot be broken in a single period of the pressure fluctuation. The value of We_{min} has to be fixed by computations, and was found to be 2.42 within an error of 0.01. For $l = d$, when drops of diameter greater than d_{max} are broken, breakage always takes place away from the centre. Therefore, at least one satellite droplet is always formed in these cases. Further, the size of the satellite drop thus formed increases as the size of the parent drop increases. It was also found that breakage was posible when $l \leqslant d$ for $d \geqslant d_{max}$. Under these conditions, highly deformed shapes are produced, suggesting the possibility of further fragmentation due to instabilities to form multiple drops.

Existing models of drop breakage in turbulent flows are largely one-dimensional in nature, have used *ad hoc* breakage conditions and assumed binary breakage. Binary breakage is not in conformity with observations. A more fundamental approach is called for to shed some light on these issues. The results obtained here qualitatively agree with observations of drop breakage in turbulent flows. Our approach enables us to determine whether a drop breaks by actually following its shape and an arbitrary breakage criterion is not necessary. It also indicates the formation of multiple daughter droplets upon breakage of a parent drop. The present approach needs further examination after relaxing some of the assumptions made.

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