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BRIEF COMMUNICATION

BUBBLE COALESCENCE IN TURBULENT FLOWS

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1. INTRODUCTION

There is considerable interest in developing means of predicting bubble-size distributions in turbulent two-phase dispersions. An understanding of the physical mechanisms determining bubble size is crucial to any detailed theory of the transfer of heat, mass and momentum between phases, and is also necessary for the framing of rules for the design of reduced-scale laboratory models to simulate bubble and droplet flows in industrial plant.

The complexity of turbulent two-phase dispersed flows necessitates analysis in statistical terms. The primary contributions to the study of such flows are those of Kolmogoroff (1949) and Hinze (1955). These authors independently suggested that the maximum size of bubble stable against breakup by the turbulence could be estimated by means of dimensional analysis based on the hypothesis that the key parameter characterizing the structure of turbulence fluctuations is the rate of energy dissipation in the flow. The purpose of the present brief communication is to point out that a similar line of argument can be applied to coalescence. It turns out that in a turbulent environment there is a minimum size of bubble stable against coalescence, as originally discovered empirically by Shinnar (1957).

Attention will be confined to the case in which the continuous phase is liquid, and it will be assumed that the intensity of agitation is sufficient to render buoyancy effects negligible, so that it is immaterial whether the dispersed phase is liquid or vapour (i.e. drops or bubbles). Detailed predictions are presented only for circumstances in which the bubble surface is effectively immobilized, e.g. as a result of surface-active contaminants.

No account is taken of the way in which the presence of the dispersed phase affects the turbulence of the continuous phase (see Lance 1979 for data on this effect); the drops or bubbles are regarded as passively acted upon by fluctuating stresses which are assumed to be the same as would obtain if the continuous phase alone were present.

Experimental studies of the factors determining bubble size have made use predominantly of either the stirred-tank configuration, in which a two-phase mixture in a closed vessel is agitated by a rotating or reciprocating paddle, or turbulent two-phase dispersed flow in pipes. These are the arrangements envisaged in the following discussion.

2. DESCRIPTION OF THE TURBULENCE

The structure of the turbulence will be described in the manner originally proposed by Kolmogoroff (1941 a, b, c) and subsequently discussed by Batchelor (1953). The turbulence eddies are considered to fall broadly into three categories, referred to, in decreasing order of size, as the energy-containing range, the inertial subrange and the viscous range. Energy is envisaged as cascading from small to large wavenumber components of the flow, i.e. from the energy-containing range to the dissipative structures of the viscous range via the eddies of the inertial subrange.

The velocity and length scales of the energy-containing eddies are identified with the turbulence intensity, u, and the integral scale, l (i.e. the integral of the two-point velocity

correlation function: Batchelor, 1953). If these two quantities are unknown for the system of interest, crude estimates may be made. For a stirred tank, u is expected to be of the order of the speed of the impeller tip, while l is probably comparable with the impeller radius; for a flowing system, u is usually between 5% (for pipe flow and boundary layers) and 25% (wakes and jets) of the variation in mean velocity across the flow and l is of order 10% of the flow width (Townsend 1976).

Eddies of the inertial subrange are largely independent both of the large-scale geometry of the flow and of the small-scale dissipative eddies. Since the functional role of the inertial-subrange eddies is simply to convey energy upwards through the wavenumber spectrum, their structure is supposed to be the same for all flows with the same energy dissipation per unit mass, ϵ. The latter quantity can be measured (e.g. by plotting the torque-speed curves for the impeller in the stirred-tank arrangement, or by pressure-drop measurements in flowing systems) or alternatively estimated from the relation

$$\epsilon \sim u^3/l$$

(Batchelor, 1953). Here, as elsewhere below, the symbol $\sim$ denotes equality within a factor of order unity.

The viscous range is responsible for the final degradation of mechanical energy into heat and is hypothesized to have a universal isotropic structure which depends only on the quantity of energy which it is required to dispose of, ϵ, and the kinematic viscosity, ν, the property which is the ultimate agency of dissipation. Dimensional analysis then gives a natural length scale of $(\nu^3/\epsilon)^{\frac{1}{4}}$ for these smallest eddies.

The Kolmogoroff–Hinze prediction of maximum bubble size follows easily from the foregoing physical picture. It is supposed that a bubble of diameter d breaks up if a critical Weber number of order unity is exceeded:

$$\frac{\rho\Delta^2}{\sigma/d} > \text{const.} \qquad [1]$$

where ρ is the density of the continuous phase, σ is the surface tension and Δ is the velocity difference across the bubble. The relation [1] may be regarded as the condition for transient pressure fluctuations associated with the turbulence to overcome capilliary forces which tend to keep the bubble intact. Now, it is usually observed that bubble sizes are such that $(\nu^3/\epsilon)^{\frac{1}{4}} \ll d \ll l$, so one expects bubble breakup to be effected by eddies in the inertial subrange. These eddies have no intrinsic velocity or length scale, so the only possible expression for Δ is of the form

$$\Delta \sim (\epsilon d)^{1/3}. \qquad [2]$$

Substitution of [2] into [1] then yields

$$d_1 \sim (\sigma/\rho)^{3/5}\epsilon^{-2/5} \qquad [3]$$

where d_1 is the diameter of the largest bubble stable against breakup.

Possibly the clearest evidence for the validity of this result when coalescence, wall effects and other complications are excluded is provided by the work of Sevik and Park (1973), who injected bubbles into the irrotational core of a water jet and measured the downstream distance at which breakup by the developing turbulence occurred. Many other experimental studies have shown that d_1 or the Sauter mean diameter (which is expected to be roughly proportional to d_1) follows the trend given by [3]: see, for example, Clay (1940), Vermeulen, Williams and Langlois (1955), Calderbank (1958), Pavlushenko and Yanishevskii (1959), Rodriguez, Grotz and Engle

(1961), Sprow (1967), Mlynek & Resnik (1972) and Coulaloglou & Tavlarides (1976), all of whom carried out experiments on the stirred-tank geometry, and Baranayev, Teverovski & Tregubova (1949), Middleman (1974), Kubie & Gardner (1977) and Karabelas (1978), who studied two-phase pipe flow. Results at variance with [3] (Roger *et al.* 1956, Sleicher 1962, Paul & Sleicher 1965, Middleman 1974, Collins & Knudsen 1970) may be plausibly attributed to secondary factors such as buoyancy and the persistence of initial bubble-size distributions.

In order to extend this kind of analysis to coalescence it is necessary to derive an expression analagous to [1] describing the conditions under which two bubbles coalesce.

3. MECHANISM OF COALESCENCE

The coalescence of a pair of bubbles occurs in two stages: (i) the draining of the intervening film of continuous-phase liquid to a critical thickness, h, which is thought to be between 10 and 100 nm (Lee & Hodgson 1968); (ii) the rupture of the remaining film by a mechanism which is not understood and may involve non-continuum effects. The first of these steps is ordinarily the slower and hence determines the overall duration of the coalescence process.

A number of attempts have been made to describe the film-drainage stage mathematically (Lee & Hodgson 1968, Murdoch & Leng, 1971, Reed *et al.* 1974, Ivanov & Traykov, 1976, Traykov & Ivanov 1977, Jones & Wilson 1978) and several detailed experiments have been carried out (Scheele & Leng 1971, Kirkpatrick & Lockett 1974, Vijayan *et al.* 1975). These studies reveal that the film-drainage process cannot be described in simple terms. Among the difficulties encountered are: (i) if a realistic zero-tangential-stress boundary condition is applied to the draining film and surface tension is correctly included, a singular perturbation problem is generated which requires matched asymptotic expansions for its solution; (ii) if the bubbles collide with appreciable velocity, circulation patterns within the bubbles impose on the film interfacial shear stresses which assist drainage, greatly increasing computational difficulties; (iii) the area of the draining film varies with time: collisions at low relative velocities, being associated with initially small film areas, are more likely to lead to coalescence than collisions at high relative velocities, which involve large deformations of the bubbles and hence the rapid formation of a large-area, slowly draining film.

To avoid these complications, which would mask the straightforward nature of the subsequent dimensional analysis, simplifying assumptions are necessary. It will be supposed that the two bubbles are pressed together by a steady force, F (which will later be related to turbulent pressure fluctuations), and that drainage takes place between rigid planes. The dynamics of the intervening film is then the same as for a film trapped between two discs, as indicated in figure 1. The assumption of a steady force pressing the bubbles together is equivalent to neglecting the period of first contact prior to the attainment of a quasi-steady state. This will be valid unless coalescence is very rapid. The assumption of drainage between rigid planes (rather than free surfaces) restricts the analysis to bubbles whose surfaces have been immobilized by surfactants, or to droplets of a dispersed phase whose viscosity is much

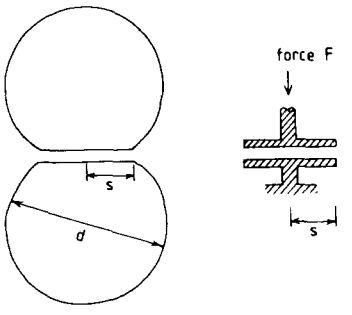

Figure 1. Model for film drainage prior to coalescence.

higher than that of the continuous phase. The analysis could, in principle, be generalized to cover the case of bubbles with mobile surfaces if one were willing to make use of numerical results for film-drainage times taken from the theoretical studies mentioned above.

Calculation of the time τ required for the films shown in figure 1 to drain to the rupture thickness h is a classical lubrication problem; it is easily shown that

$$\tau = \frac{3\pi\mu s^4}{2Fh^2}$$

where μ is the dynamic viscosity of the continuous phase, s is the radius of the film and F is the force pressing the bubbles (or discs) together. Consideration of the pressure within the deformed bubbles gives

$$F = 4\pi s^2 \sigma / d$$

so the film-drainage time may be expressed as

$$\tau = \frac{3}{32\pi} \mu F (d/\sigma h)^2. \qquad [4]$$

4. MINIMUM BUBBLE SIZE IN TURBULENT FLOW

In order to apply the result [4] to turbulent dispersions, an expression for F is needed. If it is assumed, as in the Kolmogoroff–Hinze theory, that the eddies responsible for coalescence belong to the inertial subrange, then dimensional analysis indicates that $F \sim \rho\Delta^2 d^2$, or, using [2],

$$F \sim \rho\epsilon^{2/3} d^{8/3}$$

so that [4] can be written

$$\tau \sim \frac{3}{32\pi} \mu\rho\epsilon^{2/3} d^{8/3} (d/\sigma h)^2. \qquad [5]$$

In an agitated dispersion, bubbles are continually being brought together and then moved apart by turbulent fluctuations. Let T denote the characteristic timescale of a two-bubble encounter, i.e. the length of time during which the situation shown in figure 1 persists before the two bubbles are moved apart again by the turbulence. Again assuming that eddies of the inertial subrange are the cause of bubble movement, dimensional analysis yields

$$T \sim (d^2/\epsilon)^{1/3}. \qquad [6]$$

A simple criterion for coalescence is $\tau < T$. In other words, unless the intervening film thins down to the critical rupture thickness h in the time available before bubbles are separated again, coalescence does not occur. Combining [5] and [6] we obtain the result that coalescence is impossible unless $d < d_2$ where

$$d_2 \sim 2.4 \left(\frac{\sigma^2 h^2}{\mu\rho\epsilon}\right)^{1/4}. \qquad [7]$$

This is the central result of the present paper.

Since arguments based on Kolmogoroff's hypothesis are statistical in nature, it would be more accurate to say that bubbles whose diameter exceeds d_2 are much less likely to coalesce

than smaller bubbles in the dispersion; similarly, bubbles slightly below the threshold size will not coalesce as easily as very small bubbles. The formula [7] is to be interpreted merely as a rough estimate of the size of the smallest bubble stable against coalescence.

Two aspects of the theory require further comment.

First, it has been assumed that drainage takes place between planes steadily pressed together for a period determined by the lifetime, T, of turbulent fluctuations. If this picture is to be plausible it is necessary to show that the natural rebound time for colliding droplets, t_0, is substantially less than T, so that the droplets promptly separate when the turbulence eddies are no longer favourably configured. The demonstration is straightforward. The natural vibration frequency of a droplet (Sevik & Park 1973) is of order

$$\left(\frac{\sigma}{\rho d^3}\right)^{\frac{1}{2}}$$

so, making use of [6],

$$\frac{t_0}{T} \sim \left(\frac{d}{d_1}\right)^{5/6}.$$

Hence, provided $d_2 \ll d_1$, self-consistency is assured (but see below).

Secondly, it is necessary to check that the predicted d_2 is appreciably larger than the Kolmogoroff microscale, $(\nu^3/\epsilon)^{\frac{1}{4}}$. Directly from [7],

$$d_2 \Big/ \left(\frac{\nu^3}{\epsilon}\right)^{\frac{1}{4}} = 2.4\frac{(\rho\sigma h)^{1/2}}{\mu}$$

Taking $h = 10^{-7}$ m and inserting physical properties appropriate to water at ambient temperature, the ratio is 6.4. This is perhaps just sufficiently large to justify our assumption that coalescence is controlled by eddies of the inertial subrange; for a more viscous continuous phase, however, the theory presented here will need modification.

5. THE COALESCENCE-PREVENTION REGIME

Shinnar (1957, 1961) and Shinnar & Church (1960) set up experiments in which the dominant factor determining droplet size was not breakup but the prevention of coalescence by the action of turbulence. A series of measurements was performed on dispersions of droplets in a stirred tank, to which a protective colloid had been added (immobilizing the droplet surfaces). It was found possible to form a dispersion in which no coalescence at all occurred at constant stirrer speed. This was proved by injecting dye into a single drop and observing that none spread into other droplets over a period of several hours. If, however, stirrer speed was reduced, coalescence took place at once until a new, larger stable droplet size was established, whereupon no further intermixing, as tested by the dye tracer, occurred. For these dispersions, stabilized against coalescence by the agitation imparted by the stirrer, the mean droplet size was found experimentally to be proportional to $\epsilon^{-1/4}$, in precise agreement with [7] above. It was also found that if ϵ exceeded a critical value, which will be denoted here by ϵ_0, it was no longer possible to prevent intermixing and the more usual variation of droplet size as $\epsilon^{-2/5}$ then appeared (see [3]). The observed behaviour is depicted by the solid line in figure 2. Shinnar refers to the region $\epsilon < \epsilon_0$ as the coalescence-prevention regime.

Not only does [7] give the correct trend of Shinnar's data with ϵ in the coalescence-prevention regime, it also predicts satisfactorily the absolute size of droplets, as will now be shown. Shinnar did not measure ϵ directly, so it is necessary to estimate this parameter in the way indicated in section 2 above: if D is the impeller diameter and N the number of

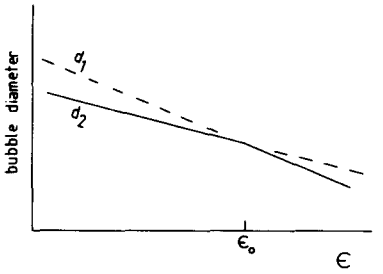

Figure 2. Variation of d_1 and d_2 with ϵ (logarithmic plot).

revolutions per unit time, we take $u \sim \pi DN$, $l \sim D/2$ and $\epsilon \sim u^3/l = 62D^2N^3$. For a 5-inch paddle at 156 rpm this method yields $\epsilon = 17.6\ \mathrm{m^2/s^3}$. The physical properties appropriate to Shinnar's dispersions are $\mu = 360\ \mu\mathrm{N\ s/m^2}$, $\sigma = 0.0385\ \mathrm{N/m}$, $\rho = 964\ \mathrm{kg/m^3}$. Finally we take $h = 0.1\ \mu\mathrm{m}$, since it seems unlikely that the thinner films sometimes observed in quasi-static coalescence experiments could survive in the highly agitated environment of a stirred tank. Putting these figures into [7] gives $d_2 = 95\ \mu\mathrm{m}$, which is to be compared with an experimental value of $128\ \mu\mathrm{m}$ (Shinnar 1961). In view of the crude manner in which ϵ has been estimated, the agreement may be regarded as encouraging.

Another way of testing [7] against Shinnar's data is to consider the magnitude of ϵ_0. Experimental evidence (Hinze 1955, Kubie & Gardner 1977) suggests that [3] becomes a true equality if a numerical factor of 0.725 is included on the r.h.s. It follows that

$$\epsilon_0 = 0.0034 \left(\frac{\sigma^2 \mu^5}{\rho^7 h^{10}}\right)^{1/3}. \qquad [8]$$

This formula has limited predictive value, however, because the numerical coefficient on the r.h.s. is extremely sensitive to the assumption that the energy dissipation is uniform throughout the dispersion. Suppose, for example, that in a stirred tank there are regions in which the turbulence intensity falls to a third of its value near the impeller, implying a local reduction in ϵ by a factor of 27 (ignoring any possible variations in l). Coalescence will be favoured in these quiescent regions, and it is readily shown that ϵ_0 will be smaller than the value given by [8] by a factor of 243. For this reason, it is probably more useful to regard ϵ_0 as a parameter which must be determined experimentally, and then to use [8] to estimate the effective value of h. For one of Shinnar's dispersions, d_1 and d_2 became equal to about 440 rpm, which implies $\epsilon_0 = 394\ \mathrm{m^2/s^3}$. Assuming that a three-fold variation in turbulence intensity did, in fact, occur within the test vessel as discussed above (although there is no experimental evidence on this point), it then follows from [8] (with the numerical coefficient reduced by the factor of 243) that $h = 0.24\ \mu\mathrm{m}$, a plausible result.

It seems that the present treatment gives a fairly satisfactory explanation for Shinnar's data. Shinnar (1961) himself attempted to explain the dependence of droplet size on ϵ in the coalescence-prevention regime by postulating that two approaching droplets cannot coalesce if the kinetic energy of collision exceeds the work necessary to affect their reseparation. To estimate this latter quantity he made use of expressions derived by Bradley (1932) and Derjaguin (1934) for the force between two macroscopic spheres which arises from the van der Waals attraction between individual constituent molecules. Valentin (1967) has drawn attention to the shortcomings of such an argument. The present analysis has the advantage of being based on familiar concepts of fluid mechanics.

It would appear from figure 2 that two-phase dispersions for which $\epsilon < \epsilon_0$ differ in an essential way from those where $\epsilon > \epsilon_0$. If $\epsilon < \epsilon_0$ (so that $d_2 < d_1$) bubbles with diameters lying between d_2 and d_1 are too large to coalesce and also too small to be broken by the turbulence; since these bubbles are stable, the bubble-size distribution must be determined largely by initial

conditions, and hysteresis phenomena are to be anticipated if ϵ is varied. If $\epsilon > \epsilon_0$, on the other hand, a true dynamic equilibrium is possible between coalescence and breakup. (It should be remarked that the present coalescence theory is not strictly applicable when $\epsilon > \epsilon_0$, because then $t_0 > T$, as discussed in Section 4.) The significance of the existence of two distinct types of dispersion has been emphasized by Church & Shinnar (1961). Shinnar's experiments suggest that for dispersions in which the continuous phase is near ambient temperature, ϵ_0 is of order 10^2–10^3 m^2/s^3. Such values are commonly encountered both in laboratory experiments and industrial plant, so one should expect to see both kinds of dispersion in practice.

6. OTHER COMPARISONS WITH EXPERIMENT

The dependence of d_2 on σ, ρ and μ cannot be deduced directly from [7] because the variation of h with physical properties is unknown. It is nevertheless of interest that if h is independent of μ, it follows from [7] that the diameter of the largest coalescing bubble varies as $\mu^{-1/4}$, so one should expect average bubble size in an agitated dispersion to decrease only slowly as μ increases.

The predicted weak dependence of bubble size on continuous-phase viscosity seems to be in agreement with some work of Calderbank (1958), who found in a series of experiments on liquid–liquid and gas–liquid dispersions that correlations for bubble size were improved by inclusion of a factor $\mu^{-1/4}$. However, Vermeulen *et al.* (1955) in a similar set of experiments found that bubble size tended to increase slightly with increasing continuous-phase viscosity. Calderbank (1956) attributed this discrepancy to Vermeulen's failure to take viscosity into account when inferring ϵ (which was not directly measured) from the known stirrer speed. Pavlushenko & Yanishevskii (1959), in yet another set of stirred-tank experiments, found, in agreement with Calderbank, a weak inverse dependence of bubble size on continuous-phase viscosity.

Thus, although there is some support for the kind of viscosity dependence given by [7], the experimental evidence is too contradictory to allow any firm verdict.

7. DISCUSSION AND CONCLUSIONS

It has been shown that the Kolmogoroff–Hinze theory can be extended to coalescence, and that the result takes a simple form [7] in the case where bubble surfaces are immobile. The derived formula provides an explanation for the results of Shinnar's important experiments on the coalescence-prevention regime.

The treatment amounts to little more than dimensional analysis guided by a simplified physical picture of the coalescence mechanism, in the same spirit as the original Kolmogoroff–Hinze breakup theory. The true complexity of coalescence events has been disregarded in favour of an easily understood idealization, just as bubble breakup is, in reality, substantially more complicated than envisaged in the Kolmogoroff–Hinze description (van't Riet & Smith 1973, Stephenson 1974, Park & Blair 1975). Furthermore, no attempt has been made to discuss kinetic aspects of the coalescence and breakup processes, although this is clearly necessary to obtain bubble-size distributions (Valentas & Amundson 1966, Jakubowsky & Sideman 1976, Narsimhan *et al.* 1979). In spite of these limitations the approach set out above may prove a useful aid to the interpretation of data and the formulation of two-phase modelling rules.

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