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Break-up of bubble clusters in turbulent flow –Theory

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Abstract

The behaviour of bubble clusters in turbulent conditions has been studied theoretically. The cluster behaviour was modelled based on concept drawn from the related field of bubble breakup. It was assumed that the bubbles were bridged by particles, so the cohesive strength was determined by the capillary force between the bubbles and the particles. Two different theories were investigated for the disruptive force from the turbulent liquid: the shear rate hypothesis of Camp and Stein (1943), and the turbulent fluctuation model arising from Kolmogorov's theory of isotropic turbulence (Kolmogorov, 1941). It was found that neither method is applicable in the fragmentation stage. However, in the equilibrium stage, an equation derived from Camp and Stein's theory was more realistic than that obtained using Kolmogorov's equation.

Keywords: Flotation, bubble clusters, bubble size

Introduction

In froth flotation, air bubbles are introduced to a slurry of ground particles in water where they collide with the suspended particles. The hydrophobic particles attach to the rising bubbles and are **lifted** up to the surface of slurry. Most flotation models are based on the collection of particles by individual bubbles. However, there are indications in the literature suggesting that hydrophobic particles are recovered with the attachment of multiple air bubbles (i.e. Schulze, 2004). Indeed, the authors recently observed that bubble aggregates could easily

form in a mechanical flotation cell (Ata and Jameson, 2005). Each bubble seemed to be held to the others by bridging particles that were attached to one or more bubbles and the shape and structure of the clusters seem to change significantly with the hydrophobicity of the particles.

Following this observation, the authors carried out the first systematic study on the clusters behavior in a well-controlled turbulent environment (Chen et al. 2015). An apparatus was used in which bubble clusters could be formed and rise into respective flow fields. The changes in the shape and size were monitored visually. Clusters properties were studied at various collector and frother concentrations, and flow conditions. It was seen that the clusters were relatively fragile, and could be disrupted by too high a level of turbulence in the cell. The experimental results indicated that a critical impeller speed exists, separating the behaviour of the bubble clusters into two stages: fragmentation and equilibrium stages. In the fragmentation stage, at low impeller speeds, the clusters were loose and filamentous, and as the energy input increased, they ruptured and re-formed (Chen et al. 2015). In the second stage, above a critical impeller speed, dense clusters formed whose size was relatively insensitive to the energy input. Apart from the maximum size of the clusters, their shape factor (SF) was also examined, which was found from the measured perimeter and the projected area of the clusters (Chen et al. 2010). It was found that although the clusters became larger at higher collector concentrations, the shape factor declined slightly, indicating a more open structure. Although these studies give an idea about the behavior of clusters in flotation cells, at present there is a lack of understanding of the forces that hold clusters together, and the hydrodynamic conditions that are most favourable to their creation.

A field where aggregates are formed is the flocculation process for water and wastewater treatment. In this process fine particulates are caused to flocculate by the addition of reagents with charged sites that form bridges between particles in an analogous fashion to cluster

formation in flotation, where the particles form bridges between bubbles. The composition of flocs does not necessarily remain the same. A continual process of growth and destruction takes place, with the ultimate diameter occurring when the rate of formation of the flocs equals the rate of destruction. The steady-state floc size is closely related to the floc strength and empirical expressions are commonly used to predict the floc strength rather than theoretical approaches due to the complexity involved in the process. The maximum floc size in a shear flow is expressed empirically as $d_{\max} = CG^{-m}$ where G is the average velocity gradient, C is the strength coefficient and m is the strength constant (Jarvis et al., 2005; Bache, 2004; Parker et al., 1972). We used this equation to study the bubble clusters breakup (Chen et al., 2010), however the equation did not provide a satisfactory agreement with the experimental results.

This paper presents a simple theoretical analysis to give insight into the behavior of clusters under shear flow. By drawing an analogy between bubble clusters and bubble/droplet breakup, a model based on force balance is proposed to predict the maximum stable size of a bubble cluster in turbulent conditions. The cohesive capillary force and the detachment forces of hydrodynamic origin are balanced in the model. An important variable in the model is the shear rate used in the calculation of the dynamic detachment force. It should be borne in mind that forces acting on bubble clusters are more complicated in nature because clusters contain both bubbles and particles and the forces within clusters are not in an ordered direction. As a starting point, a simple case, such as two bubbles joined with one particle, were considered.

Description of theory

When a bubble is immersed in a turbulent flow, it experiences turbulent stresses including viscous stresses and dynamic pressures, which tend to deform or break the bubbles (Hinze,

1955). The same type of stresses could be considered to act on a cluster of bubbles held together by bridging particles. In the case of the bubble, the force that resists breakup is due to the surface tension of the bubble. In the case of the cluster, it is the internal cohesive forces arising from the capillary force between the bubbles and the bridging particles. When the disruptive forces exceed the internal force, the bubble cluster is broken up.

By comparing the disruptive and internal cohesive forces, a model can be proposed to predict the breakage of bubble clusters. For simplicity, we only consider the case that two air bubbles are connected with each other by a hydrophobic particle to avoid the effect of structural factors on the stability of bubble clusters. We will employ the following equation to express the maximum capillary force between a particle and a bubble, from Nguyen (2003) in the form:

$$F_c = 2\pi R_p \sigma \sin^2(\theta/2) \quad [1]$$

where R_p is the radius of particle, σ is the gas-liquid surface tension and θ is the contact angle (see Figure 1).

The disruptive dynamic pressure force is equal to the dynamic pressure multiplied by the area where the pressure applies, which can be taken to be the area of cross-section of an individual bubble cluster, πR_c^2 , where R_c is the radius of cluster. Thus, the dynamic pressure force can be expressed as:

$$F_d = \pi R_c^2 \rho_l \overline{u_d^2} / 2 \quad [2]$$

where ρ_l is the density of fluid, $\overline{u_d^2}$ is the mean square velocity difference over a distance equal to the diameter of the bubble cluster, d_c .

Figure 1 Bubble cluster in turbulent flow. Dynamic pressure force due to the flow velocity difference at opposite ends of the cluster causes the cluster deformed and breakup (a). At steady-state this force is balanced by the capillary force which is function of the particle radius R_p , the gas-liquid surface tension σ and the contact angle θ (b).

Two assumptions can be made to predict the value of $\overline{u_d^2}$. Firstly, it will be assumed that the velocity difference can be calculated by relating it to the shear rate. If a cluster is in a uniform shear field, the velocity difference at opposite ends of a cluster is $\Delta u_d = G d_c$, where d_c is the cluster diameter G is the shear rate. The shear rate can be calculated from the equation of Camp and Stein (1943) as: $G = \sqrt{\varepsilon / \nu}$, where ν is kinematic viscosity, ε is energy dissipation per unit mass of fluid, which is given by $\varepsilon = P_0 N^3 D^5 / V$, where P_0 is the impeller power number, N is the impeller speed, D is the impeller diameter, and V is the volume of the liquid in the stirred tank. By substituting all parameters, one can obtain:

$$\left(\overline{u_d^2} \right)_C = \varepsilon d_c^2 / \nu \quad [3]$$

where the subscript C of $\overline{u_d^2}$ denotes that this expression uses the Camp and Stein expression.

By substituting Equation [3] into Equation [2], the dynamic pressure force can be modified to:

$$\left(F_d \right)_C = \pi \rho_l \varepsilon d_c^4 / 8 \nu \quad [4]$$

The value of $\overline{u_d^2}$ can also be obtained according to Kolmogorov's theory of local isotropy (Kolmogorov, 1941). It is assumed that at sufficiently high Reynolds numbers (>10000) the micro-scale components of the turbulent velocity fluctuations are isotropic and therefore independent of the main flow and the generating mechanism of the flow. In the case of a droplet in an isotropic turbulent flow, that is much larger than the micro-scale, the mean square of the relative flow velocity over a distance equals to the diameter of droplet d is independent of viscosity, which can be expressed as (Shinnar, 1961):

$$\left(\overline{u_d^2}\right)_K = 2\varepsilon^{2/3} d_c^{2/3} \quad [5]$$

where the subscript K of $\overline{u_d^2}$ denotes that this expression uses Kolmogorov's theory. An analogy can be drawn with the case of a cluster, where the cluster size is much larger than the microscale, in which case d_c is the diameter of the cluster.

By substituting Equation [5] into Equation [2], the dynamic pressure force can be modified to:

$$(F_d)_K = \pi\rho_t\varepsilon^{2/3} d_c^{8/3} / 4 \quad [6]$$

and it can be observed that the values of F_d depend in different ways on the key variables ε and d_c .

For real systems, one bubble may be connected with many particles so that many joints may be broken up at the same time. If the average number of broken contacts of clusters is N_c it would be reasonable to assume that for stability, the dynamic pressure force should be balanced by N_c times the capillary force. By balancing of Equations [4] and [6] with N_c times the attachment force expressed in Equation [1], the maximum stable size of bubble clusters in turbulent conditions can be obtained:

$$(d_c)_c = 2 \left(N_c R_p \sigma \sin^2(\theta/2) \nu \right)^{1/4} \rho_l^{-1/4} \varepsilon^{-1/4} = 2 N_c^{1/4} \left(\frac{\rho_l \varepsilon}{R_p \sigma \sin^2(\theta/2) \nu} \right)^{-1/4} \quad [7]$$

$$(d_c)_k = \left(8 N_c R_p \sigma \sin^2(\theta/2) \right)^{3/8} \rho_l^{-3/8} \varepsilon^{-1/4} = 2.18 N_c^{3/8} \left(\frac{\rho_l^{3/2} \varepsilon}{R_p^{3/2} \sigma^{3/2} \sin^3(\theta/2)} \right)^{-1/4} \quad [8]$$

The accuracy of these equations will be tested against the experimental data obtained in well-controlled turbulent environment. The differences between two equations will be discussed.

Verification of the theory

The formation and breakup of bubble clusters were studied experimentally in an apparatus where the bubble clusters were pre-generated and then broken up by a shear flow. Clusters properties were studied at various collector concentrations (dodecylamine, DDA) and impeller speeds. The experimental details and results can be found in Chen et al. (2015) (see also Chen et al., 2010). It appeared that the clusters present two different behaviours with increasing impeller speeds: fragmentation and collision stage, which is separated by a critical impeller speed. At fragmentation stage, the breakup of clusters is highly related to the initial size of clusters while at collision stage, the size of clusters is dependent on the balance between breakup and reformation of clusters. The clusters are mainly formed by collision between particles and bubbles, bubbles and clusters, or bubbles loaded with particles. This stage is also in equilibrium, that is, where processes of cluster formation and rupture are in balance.

Table 1 shows the values of the contact angle of particles and surface tension at various DDA concentrations used in the model. The data was adopted from Smith (1963) and Castro et al. (1986). The energy dissipation rate for various impeller speeds is shown in Table 2, which was calculated according to the size of the impeller and the volume of flotation cell used in the experiments (Chen et al., 2015).

Bubble clusters in the fragmentation stage

Figure 2 shows the average diameters of bubble clusters as a function of $\rho_l \varepsilon / (R_p \sin^2(\theta/2) \nu)$, taken from the Camp and Stein formulation, at dodecylamine concentrations of 1×10^{-5} , 5×10^{-5} , 9×10^{-5} M. The experimental data at low impeller speeds, where the clusters are forming and re-forming were used in this figure. The equations in the figure, which are of the form $y = ax^b$, assume a linear fit to the data. The slopes of the fitting lines increase with increasing DDA concentrations and do not follow the $1/4$ law in Equation [7], suggesting that bubble clusters are in the fragmentation stage, where the sizes of the clusters is significantly affected by their initial size and shape. These factors are strongly related to the hydrophobicity of the particles. It was experimentally observed that the initial size of the clusters increases with increasing DDA concentrations and the shape of cluster changes from irregular to regular in this stage. The size of the clusters formed at high dodecylamine concentrations drops significantly as the impeller speed increases due to their initial branchy structure, while the clusters generated at lower DDA concentrations are more compact. In the fragmentation stage, the shear rate is insufficient to break the bubble clusters down into single bubbles. But the shear rate is able to cut off the weakest branches of the clusters. To model this stage, the cluster shape should be considered and implemented into the model. However, it is hard to find a proper shape expression to incorporate into the current model because cluster is composed of multiple air bubbles and solid particles which increases complexity. Attempts were made to integrate the fractal dimension based on image analysis into the model, but no promising results were obtained. It is even more difficult to obtain mass fractal dimension of clusters because of the mixture of bubbles and particles.

Figure 3 shows the average diameters of bubble clusters versus the dissipation term $\rho_l^{1.5} \varepsilon / (R_p^{1.5} \sigma^{1.5} \sin^3(\theta/2))$ drawn from the Kolmogorov formulation. The slopes of the lines

are similar to those in Figure 2, although the values of the coefficient are different. The fact that the slopes are similar indicates that at a given DDA concentration, both equations reflect the same rates of size reduction of clusters at a certain DDA concentration. The number of broken contacts N_c can be calculated from the value of the intercepts, and it appears that in each case, the factor a in $y = ax^b$ is close to zero for all DDA concentrations. It appears that neither Equation [7] nor Equation [8] can represent the phenomena taking place in the fragmentation stage.

Bubble clusters in the equilibrium stage

Figure 4 and Figure 5 illustrate the average diameter of cluster as a function of $\rho_l \varepsilon / (R_p \sigma \sin^2(\theta/2) \nu)$ and $\rho_l^{1.5} \varepsilon / (R_p^{1.5} \sigma^{1.5} \sin^3(\theta/2))$. The experimental data at high impeller speeds, where the clusters reach their equilibrium size were used in the verification of model. It is noted that a frother (methyl isobutyl carbinol, MIBC) at concentration of 80 ppm was added to the solutions to eliminate the impact of bubble size on the behaviour of bubble clusters.

In both cases, a power-law relation between the cluster diameter and the dissipation function on the y axis is observed, with an exponent of -0.25 approximately, which is in agreement with the predictions of both Equations [7] and [8].

The main differences between the two simulations are the values of the coefficients $2N_c^{1/4}$ and $2.18N_c^{3/8}$ in Equations [7] and [8], respectively. These coefficients are functions only of the number of active contact points N_c , that are just able to hold the clusters together against the action of the disruptive forces.

Using the best-fit values displayed in Figures 4 and 5, the numbers of broken contacts are calculated to be 4.3 and 0.13 respectively. The latter figure is physically unrealistic, so it can be concluded that Equation [7], which uses the Camp and Stein velocity gradient theory, is more appropriate for the calculation of the cluster breakage mechanism.

The expression for the energy dissipation rate in the paper of Camp and Stein (1943) is rather simplistic. It is drawn from a general equation for the local dissipation rate in a Newtonian fluid, expressed in Cartesian coordinates, originally derived by Stokes and explained in more detail in Lamb (1932), Article 329. It is in effect a one-dimensional equation for a three-dimensional effect, and does not recognize the effect of the Reynolds number on fluid phenomena. By contrast, Kolmogorov's theory assumes that the flow is at a very high Reynolds number, where the velocity fluctuations are independent of viscosity when particles are within the inertial sub-range in turbulent conditions. The applicability of the Camp and Stein theory suggests that bulk shear stresses play an important role in calculating the velocity differences around a bubble cluster. The dissipation term in Equation [8] appears to over-estimate the effect of the turbulent liquid on the cluster. One possible explanation is that the size of the bubble cluster is too large to be counted within Kolmogorov's inertial sub-range, which is defined as the characteristic dimension r should be far smaller than scale of largest energy containing eddies L and far larger than Kolmogorov's microscale η ($L \gg r \gg \eta$). The sizes of the bubble clusters may be of the same order of magnitude as the largest eddies, which is the width of Rushton impeller in the current study, corresponding to 10 mm.

The Camp and Stein absolute velocity gradient is probably more applicable in describing the velocity difference at length scales larger than inertial sub-range in the stirred tank. Isotropic flow probably does not exist at a length scale equal to the size of clusters, especially in the region of the impeller. The properties of the flow are more related with the total energy input and liquid characteristics.

The theoretical verification also demonstrates the different behaviours of clusters at impeller speeds smaller and larger than the critical impeller speed (CIS), that forms a boundary between the fragmentation stage and the equilibrium cluster size stage. At impeller speeds lower than CIS, the forces used in this study cannot be balanced because the strength of clusters is greatly influenced by its shape, which is unknown. At high impeller speeds, the two forces used here can be balanced if approximately 4.3 contacts are connected. The diameters of the clusters relates to the dissipation group to the power of -0.25, as predicted by the theory for the equilibrium stage. In the fragmentation stage where the pre-formed clusters are gradually broken apart as the shear rate increases, there is no simple general theory. In the equilibrium regime the effect of the shape factor disappears because all clusters at this stage have similar shapes.

Conclusions

A model to describe the behaviour of clusters was developed for the first time. The model is mainly based on balance between the disruptive force that acts on clusters and the cohesive force within clusters. The disruptive force is mainly dynamic pressure force and the cohesive force is capillary force between particles and bubbles. In expressing the dynamic pressure, the velocity fluctuation of clusters was derived by two methods: one was Camp and Stain's equation (1943) and another one derived from Komogrof's theory. The Camp and Stain's equation shows a best fit to this work according to the results. In deriving the model, the effect of bubble size on bubble clusters was eliminated by adding frother into the solution. The model provides an easy way to predict the cluster size in industrial flotation cells.

Predictions from the model were compared with the experimental results where a two-stage process for cluster formation had been observed: a fragmentation stage where the clusters are filamentous and easily broken, and an equilibrium stage where the clusters are more densely

packed and resistant to rupture. A critical impeller speed (power input) at which there is a transition from one stage to the other, was identified.

In the fragmentation stage, the theoretical model is inapplicable, because the shape of the clusters plays an important role in their breakup, and the shape changes in an unknown way under the action of the dynamical forces.

In the equilibrium stage, two methods were used to model the the shear rate or dynamic stress; one is derived from an equation of Camp and Stein (1943) while the other is drawn from Kolmogorov's theory of isotropic turbulence. Each method predicts that the cluster diameter varies as the energy input to the $1/4$ power and this dependence was corroborated experimentally. Another important component of the model is the number of particle-bubble attachment points in the cluster, which is proportional to the rupture strength. The Camp and Stein method predicts that when the clusters are in equilibrium, 4.3 bridging contacts must be ruptured for the cluster to disintegrate, but the Kolmogorov model predicts that only 0.13 contacts are needed on average, which is clearly unacceptable on physical grounds. The equation recommended for the maximum stable cluster size is:

$$(d_c)_c = 2 \left(N_c R_p \sigma \sin^2(\theta/2) \nu \right)^{1/4} \rho_l^{-1/4} \varepsilon^{-1/4} = 2 N_c^{1/4} \left(\frac{\rho_l \varepsilon}{R_p \sigma \sin^2(\theta/2) \nu} \right)^{-1/4}$$

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FIGURES

Figure 1 Bubble cluster in turbulent flow. Dynamic pressure force due to the flow velocity difference at opposite ends of the cluster causes the cluster deformed and breakup (a). At steady-state this force is balanced by the capillary force which is function of the particle radius R_p , the gas-liquid surface tension σ and the contact angle θ (b).

Figure 2 The average diameters of bubble clusters in the fragmentation stage versus $\rho_l \varepsilon / (R_p \sigma \sin^2(\theta/2) \nu)$, from the theory using Camp and Stein's velocity gradient, Equation [7].

Figure 3 The average diameters of bubble clusters in the fragmentation stage versus $\rho_l^{1.5} \varepsilon / (R_p^{1.5} \sigma^{1.5} \sin^3(\theta/2))$, using the Kolmogorov approach, Equation [8].

Figure 4 The relationship between average diameter of clusters in the equilibrium stage and $\rho_l \varepsilon / (R_p \sigma \sin^2(\theta/2) \nu)$ at high impeller speeds, using Camp and Stein's velocity gradient, Equation [7]

Figure 5 The relationship between average diameter of clusters in the equilibrium stage and $\rho_l^{1.5} \varepsilon / (R_p^{1.5} \sigma^{1.5} \sin^3(\theta/2))$, using the Kolmogorov formulation, Equation [8].

Fig 1.

Fig 2.

Fig 3.

Fig 4.

Fig 5.

TABLES

Table 1. The surface tension and contact angle of particles at various DDA concentrations.

DDA Concentration (Mol/L)	Surface tension σ (N/m)	Contact angle θ
0.00001	0.067	27
0.00005	0.055	43
0.00007	0.051	47
0.00009	0.047	50
0.00012	0.044	55

Table 2. Energy dissipation at various impeller speeds.

rpm	rps	ϵ
43	0.72	0.00011
149	2.49	0.00441
257	4.28	0.02246
377	6.28	0.07095
400	6.67	0.08488
478	7.97	0.14511
550	9.17	0.22065
700	11.67	0.45489
850	14.17	0.81445