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"Bubble Bunch" phenomenon in operation of a bubble column

Research Article

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Abstract: While studying the operation of a rectangular bubble column in laboratory scale, it was observed that under certain circumstances tiny bubbles attach to larger bubbles without causing them to coalesce. In other words, bubbles with large diameters (d > 5 mm) swept tiny bubbles (d < 1 mm) in their way to the top of the column resulting in grapelike clusters of bubbles. This phenomenon was named "bubble bunch" by us and its effect on total gas holdup and interfacial area was discussed. Although it was found to have almost no affect on gas holdup, bubble bunch can increase the interfacial area up to 10% more than what is anticipated in the literature. The process of film thinning was modeled for this phenomenon and coalescence efficiency was calculated as a function of interfacial tension.

Keywords: *Bubble bunch • Bubble coalescence • Interfacial tension • Bubble columns • Interfacial area*

1. Introduction

Bubble columns are widely used in different industries and their operation and design have been the focus of many researchers for decades. Many theoretical and experimental studies have been performed in an attempt to offer a prediction of the operation of a bubble column under different conditions [1,2].

Bubble size distribution is a hydrodynamic dominating parameter in the operation of bubble columns, since this parameter along with gas holdup (ε_g) will specify interfacial area through the equation:

$$a = \frac{6\varepsilon_g}{d_{32}} \quad (1)$$

where d_{32} is Sauter mean bubble diameter. Bubble size is either measured directly through experimentation or determined by simulation. Recent codes and researchers use a distribution function instead of an averaged diameter [3,4]. Generally the bubble size distribution changes throughout the column. The coalescence and breakage of bubbles are modeled according to our results [5] and various theories in the literature [6,7]. Then, a population balance model is solved with various available techniques [8].

At the beginning of our research, the main purpose was to verify the simulation results of the population balance model. For this reason, a rectangular laboratory scale bubble column was constructed. Under different interfacial tensions, the bubble size distribution and gas holdup were measured. During the experiments performed under conditions of low interfacial tension, very tiny bubbles, generated in the formation stage, are swept by larger bubbles with which they do not coalesce thus forming a grape shaped colony of bubbles (Fig. 1). We named this phenomenon "bubble bunch" to make distinction with "bubble grapes" [9] and "bubble cluster" [10]. The tiny bubbles in the bubble bunches are never

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taken into account in the computation and measurement of gas holdup and bubble size distribution as they have negligible effects. However, they can considerably increase the interfacial area.

Although it is the first time that such a phenomenon has been reported and investigated, it is anticipated that it will be applicable in industries using any types of gas-liquid equipment. In some units such as airlift bioreactors, gas absorption, two-phase reactors, *etc.*, the formation of bubble bunch increases the interfacial area and hence the rate of mass transfer. Conversely, in unit operations such as floatation, bunches of bubbles rise more slowly compared to single bubbles resulting in increased time necessary for separation.

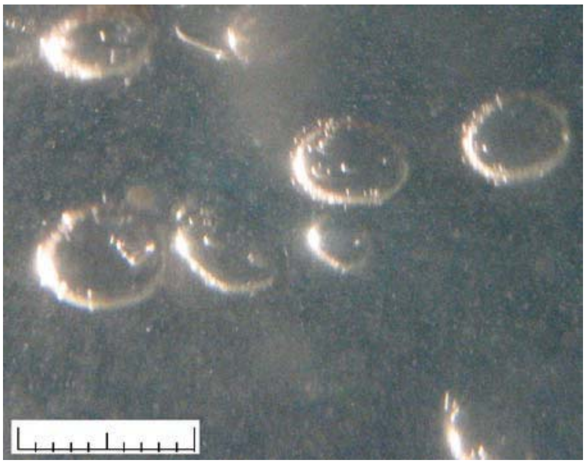

Figure 1. Formation of bubble bunch in the system containing air and butanol solution in water within a bubble column. Tiny bubbles attached to larger bubbles in the middle of picture are obvious. Scale shown on the bottom left is 10 mm.

2. Theory

When two bubbles approach each other, the formation of the liquid film between them occurs in the following stages [11]:

• Initially a hydrodynamic interaction between two bubbles takes place. This interaction is stronger at the front sections of the bubbles and leads to weak deformation of their surfaces.

• When the bubbles reach a certain small separation, called inversion thickness, the direction of the curvature in the front section of the bubbles changes and a concave lens-shaped formation called a *dimple* is formed. This stage is possible if the force acting on the bubbles is high enough to overcome the energy barrier created by the increase of surface energy during the deformation.

• At first, the dimple grows, but as a result of the outflow of liquid, it decreases and a virtually plane-parallel film is formed that afterwards thins at a constant radius.

• Thermodynamic fluctuations or outer disturbances of the film thinning process either lead to rupture of the film or to formation of black spots of much lower thickness.

• The black spots either coalesce or grow, forming a secondary equilibrium film known as a *Newton black film* (NBF). Then the spot grows until it covers the whole film area. The typical thickness of the plane-parallel films is about 200 nm, while the characteristic thickness of a NBF is about 5–10 nm. After the whole film area is taken up by the secondary film, its radius is increased until it reaches the equilibrium film radius.

Bubble coalescence is usually expressed as the product of collision frequency and collision efficiency. The efficiency of a collision is generally expressed as the process of thinning of the liquid film. It is assumed that coalescence happens when the two bubbles stay at rest in sufficient contact with each other so that the thickness of liquid film decreases to a critical value. According to Coulaloglou and Tavlarides [12], collision efficiency between two bubbles of diameter d_i and d_j may be expressed as:

$$\lambda(d_i,d_j)=\exp\left(-C\frac{\mu_l\rho_l\varepsilon_t r_{ij}^4}{\sigma_l^2}\right) \tag{2}$$

where λ is the efficiency of collision, μ_l , ρ_l and σ_l are viscosity, density and surface tension of liquid phase, respectively. ε_t is the turbulence energy dissipation rate per unit volume of continuous phase and C is an empirical constant. r_{ij} is the equivalent radius of collision defined as:

$$r_{ij}=\frac{d_i d_j}{d_i+d_j} \tag{3}$$

In Eq. 2, it is assumed that both bubbles have stagnant surfaces. Based on the above model, a similar model is proposed by Prince and Blanch for bubbles with moving surface [6]. The efficiency of collision is expressed in required time for coalescence (τ_{ij}) and contact time(τ_{ij}):

$$\lambda(d_i,d_j)=\exp\left(-\frac{t_{ij}}{\tau_{ij}}\right) \tag{4}$$

The coalescence time and contact time are:

$$t_{ij}=\left(\frac{r_{ij}^3\rho_l}{16\sigma_l}\right)^{1/2}\ln\frac{h_{f,0}}{h_{f,c}} \tag{5}$$

and

$$\tau_{ij}=\frac{r_{ij}^{2/3}}{\varepsilon_t^{1/3}} \tag{6}$$

where $h_{f,0}$ is initial thickness and $h_{f,c}$ is the critical thickness of liquid film. Substituting t_{ij} and τ_{ij} from Eqs. 5 and 6 into Eq. 4 will give an equation similar to Eq. 2 as follows:

$$\lambda(d_i,d_j)=\exp\left(-C'\frac{\rho_l^{1/2}\varepsilon_t^{1/3}r_{ij}^{5/6}}{\sigma_l^{1/2}}\right) \quad (7)$$

The constant $C'(=\frac{1}{4}\ln\frac{h_{f,0}}{h_{f,c}})$ may be measured by experiments.

To derive a correlation for the critical film thickness, consider the bubbles shown in Fig. 2. Cylindrical coordinates are chosen for the system. The large bubble is illustrated with a plane while the tiny bubble is a sphere. The bubble is sufficiently small that it may be assumed to be spherical. This is equivalent to assuming that the Bond number:

$$N_{Bo}=\frac{\Delta\rho g R_b^2}{\sigma^\infty}<<1 \quad (8)$$

Here $\Delta\rho$ is the magnitude of the density difference between the bubble phase and the continuous phase, g the magnitude of the acceleration of gravity, R_b the radius of the small bubble, and σ^∞ the equilibrium interfacial tension. In the experiments done in the present work, the Bond number for small particles is more sensitive to the changes in interfacial tension than to gravity and varies from 0.03 to 0.12. Because the values is well below 1, the effect of gravity on bubble shapes can be ignored.

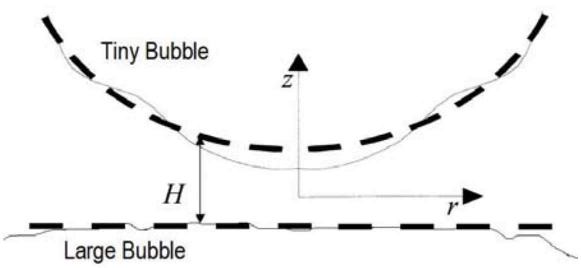

Figure 2. A sketch of a tiny bubble adjacent to a large bubble.

The distance between two bubbles can be approximated by the following equation:

$$H=h_{f,0}+\frac{r^2}{R_b} \quad (9)$$

Then the relations for two bubbles under external force obtained by Valkovska *et al.* [13] reduces to:

$$h_{f,c}^3=\frac{3}{160}\left(\frac{R_bA_H}{\pi\sigma}\right) \quad (10)$$

where A_H is the Hamaker constant defined as:

$$A_H=\pi^2C\upsilon_1\upsilon_2 \quad (11)$$

where υ_1 *and* υ_2 are the number of atoms per unit volume in the two bodies and C is the coefficient in the atom-atom pair potential.

Inserting Eq. 10 into Eq. 7, and defining m as the ratio of two bubble diameters the following simplified equation for coalescence efficiency is derived:

$$\lambda=\left(\frac{3A_HR_b}{160\pi h_{f,0}^3\sigma}\right)^{\left(\frac{\rho_l^{1/2}\varepsilon_t^{1/3}\left(2\frac{m}{m+1}R_b\right)^{5/6}}{12\sigma^{1/2}}\right)} \quad (12)$$

Introducing the coefficients $k_1=\frac{160\pi h_{f,0}^3}{3A_HR_b}$ and $k_2=\frac{1}{12}\rho_l^{1/2}\varepsilon_t^{1/3}\left(\frac{2m}{m+1}R_b\right)^{5/6}$ one finally obtains:

$$\lambda=(k_1\sigma)^{-\left(\frac{k_2}{\sigma^{1/2}}\right)} \quad (13)$$

3. Experimental Setup

A rectangular bubble column was made of float glass with dimensions of 20×40×120 centimeters (as shown in Fig. 3). The sparger was made of a 15×30 cm stainless steel 304 plate with a thickness of 4 mm and perforated with fine holes of 500 microns to generate uniform bubbles. The holes are arranged in 4×9 rows with a pitch of 2 cm.

The sparger was screwed to the end of a pyramid shape distributor, and the entire assembly was placed at the bottom of bubble column so that there was no liquid circulation below the sparger.

Air was compressed in a small compressor (provided with a built in air filter) made by GAST Co. Ltd. and injected as the gas phase at ambient temperature (25˚C) and 0.4 barg. The gas volume rate was adjusted with a needle valve and a KDG 1100 rotameter to 10 L min^{-1}. All valves were provided by ARYA Co.

Experiments were carried out with three different solutions: deionized water, aqueous methanol 10.8% by volume, and aqueous butanol 5% by volume.

Methanol and ethanol were of ultra-pure grade and products of Shimigostar.

Deionized water was provided by Bahrezolal Co. (conductivity < 2 micro siemens).

Gas hold-up was measured directly, and bubble size distribution determined by photographs with an A100 Sony 10 Mega Pixel digital camera with shutter speed of 1/1250 second. For tracking the changes in bubble size distribution, two sections were selected along the column.

The first section near the sparger was used to calculate the input bubble size distribution, and the second was near the top (at 4/5 height of the column) to avoid high turbulence at the top of column. The photos from the latter section were used to calculate the bubble size distribution at the exit of column. Moreover, the photo shown in Fig. 1 is also taken at the higher point because at lower sections the bubble bunch has not been completely formed. The photos were analyzed with GSA ImageAnalyser v 2.5.2 software developed by Bansemer & Scheel GbR.

For each solution, interfacial tension was measured with an IFT700 surface tension determination system of VINCI® Technologies. The measurement is based on recording drop shape factor using a special camera and calculating the interfacial tension with specific software supported with the IFT700.

The average value for 10 tests was considered as the mean interfacial tension of each solution at 25˚C.

The liquid viscosity was measured with a capillary viscometer (Ostwald viscometer) of Petrotest® at 25˚C. The results of measurements are shown in Table 1.

Table 1. Physical properties of different solutions used in the experiments

Solution	Viscosity (cP)	Interfacial tension (dyne/cm)
Water (deionized)	1.03	71.45
Methanol 10.8% v/v	1.09	54.99
Butanol 5% v/v	1.65	41.96

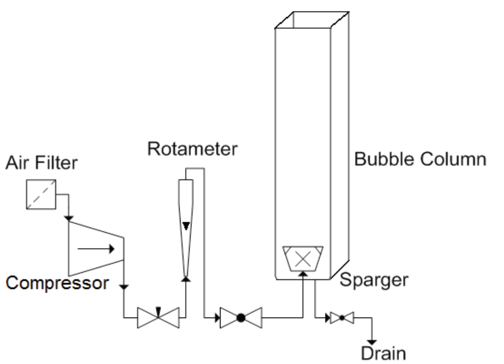

Figure 3. A schematic of the setup and bubble column used in the study.

4. Results and Discussion

The physical properties of different solutions used in the experiments are listed in Table 1. Butanol solution has the lowest value of surface tension and a viscosity similar to water. But the viscosity of methanol solution is almost equal to water.

When the coalescence efficiency obtained from Eq. 12 is plotted *vs.* bubble size and interfacial tension, a decrease of λ with decreasing interfacial tension is observed. An example of such a plot can be seen in Fig. 4. The parameters applied in this figure are $A_H = 10^{-20}$ J [14], $\varepsilon_t = 2.5$ W m^{-3} [15], $h_{f0} = 500$ nm, $\rho_l = 1$ g cm^{-3}.

Thus, tiny bubbles are kept in touch with the larger bubbles, and it was observed that large bubbles carry them for a long distance (free of charge!) without any coalescence.

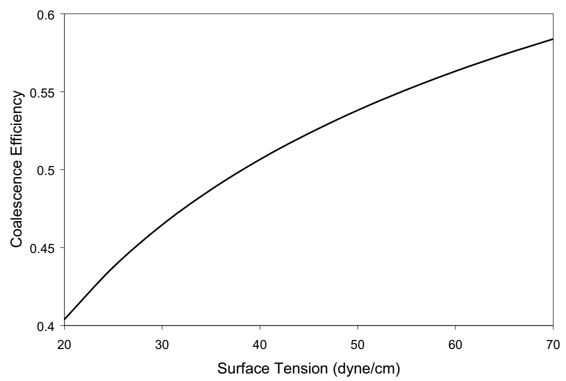

Figure 4. Coalescence efficiency *vs.* surface tension, plotted for a bubble of 0.5 mm in diameter in touch with a bubble of 5 mm in diameter.

Since this is the first observation of the bubble bunch phenomenon, it was thought that decreasing rates of coalescence due to decreases in interfacial tension are the main reason these bunches form. Hence, a theory was developed to predict the variation of coalescence efficiency with interfacial tension. The resulting equations predicted that decrease of interfacial tension will result in a moderate decrease in coalescence efficiency. This was the main link between the developed theory and our observations.

According to our observations, the bubble bunch lasted throughout the column. To investigate the actual life of the bunch, a longer column is required which was not possible at that time.

The formation of bubble bunches was not observed in water and methanol solution but only in butanol solution. This is due to the fact that the interfacial tension of butanol solution is much lower than methanol solution.

The bubble size distribution function was measured in the bottom and top of the column with conventional image analysis techniques described previously. The results are shown in two different plots: one in terms of number fraction (Fig. 5) and the other in terms of volume fraction (Fig. 6). Number fraction is defined as the fraction of the number of bubbles in each size group in accordance to the total number of bubbles counted (here 300).

From Fig. 5 it can be seen that small bubbles which initially exist at the bottom of column disappear in the top section due to coalescence, and bubble size distribution shifted slightly to the larger sizes. This is more obvious in Fig. 6 where the Sauter mean diameter is calculated to have changed from 5.16 mm to 5.52 mm (7% increase).

So the "bubble bunch" effect is never accounted in calculation of interfacial area. Although the change in volume of large bubbles due to this phenomenon is about 1%, which is negligible, the change in the interfacial area is more important; and it may increase up to 10% based on analysis of more than 200 photographs of the performed experiments. The result of this analysis is shown in Fig. 7 where a plot of interfacial area considering bubble bunch effect *vs.* interfacial area neglecting this effect is drawn.

In fact this figure represents the experimental (actual) interfacial area *vs.* theoretical (calculated) interfacial area. The diagonal line (y=x) is also plotted to show more clearly the deviation of actual values from expected calculated values.

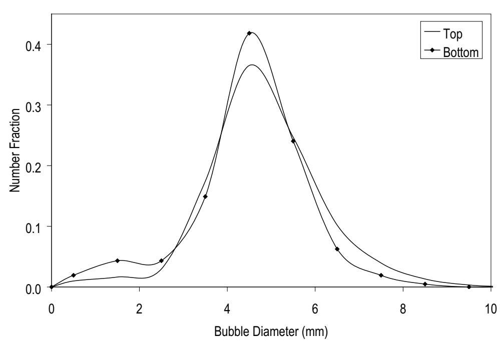

Figure 5. A comparison between bubble size distribution in term of number fraction in bottom and top of column experimentally obtained for butanol solution.

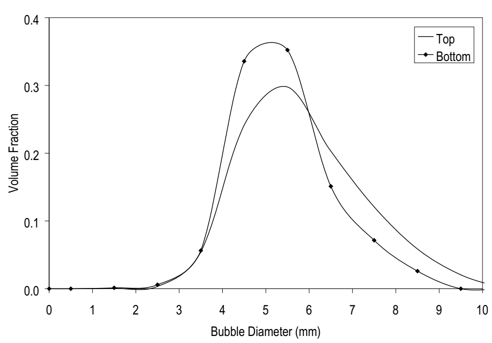

Figure 6. A comparison between bubble size distribution in term of volume fraction in bottom and top of column experimentally obtained for butanol solution.

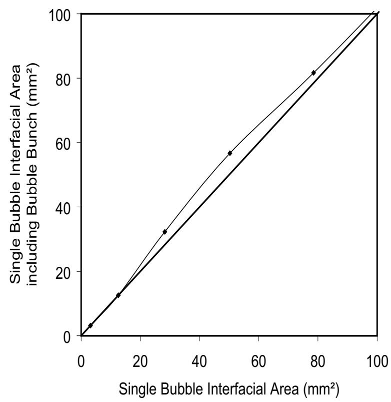

Figure 7. Interfacial area considering bubble bunch effect *vs.* interfacial area neglecting bubble bunch effect.

5. Conclusion

The effect of surface active materials was studied in a rectangular, laboratory scale bubble column. It was observed that in low interfacial tension media, tiny bubbles present in the continuous phase tend to attach to larger bubbles without coalescence to form a grape-liked structure named bubble bunch. An equation for critical film thickness was derived based on a theoretical model which showed that a decrease in interfacial tension decreases the critical film thickness thus increasing the stability of the bunch and decreasing the coalescence efficiency which is thought to be the main reason for formation of bubble bunch.

In addition, it was shown that these bunches have almost no effect on bubble size distribution expressed as volume fraction; they can alter interfacial area considerably. Experiments show a deviation of interfacial area up to 10% from its expected value as a result of bubble bunch formation. This is justified implicitly by the decreased coalescence efficiency in low interfacial tensions anticipated by the foregoing theory.

Tiny bubbles are mainly formed at the sparger during the bubble formation at low interfacial tensions, and the rate of tiny bubble formation increases with decreasing interfacial tension.

Moreover, the formation of bubble bunches in low interfacial tension is independent of the bubble size formed at the sparger.

At high superficial gas velocities, the number of very tiny bubbles considerably increases resulting in the formation of a hazy liquid which makes photography impossible. In addition, with the available digital camera and image processing software, it was not possible to track smaller bubbles than those seen in Fig.1.

Hence, it is suggested that more precise experiments should be performed in order to reveal the effect of this phenomena on different aspects of operation of a bubble column.

It is anticipated that formation of bubble bunches will have different effects on different industries. For example, while increasing the rate of mass transfer in bubble column reactors (a desirable effect); it may increase separation time in floatation units (which is an undesirable effect). To prevent the formation of bubble bunches, very low interfacial tension or formation of large bubbles at the sparger must be avoided.

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