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# EXPERIMENTAL HYDRODYNAMICS OF INTERACTING GAS BUBBLES AND OIL DROPLETS: ATTACHMENT PHENOMENA

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**Abstract.** *The attachment phenomena of gas bubbles to oil droplets was successfully evaluated in the present work, aiming the elucidation of the interactions found in several industrial applications. Pure nitrogen and colored corn oil acted as the dispersed phases, whereas distilled water was employed as the medium. The hydrodynamics of single bubbles in free gravitational motion was evaluated by experimental high speed recordings of the flow, allowing the estimation of the mean bubble diameter, aspect ratio and terminal velocity, in addition to the calculation of the governing dimensionless parameters Reynolds and Eötvös numbers. The experimental set-up in the Multiphase Flow Research Center (NUEM-UTFPR) allowed the controlled generation of monodispersed single gas bubbles and oil droplets. The attachment phenomena observed for individual bubble and droplet with diameters of, respectively, 3.11 and 5.8 mm consisted in the penetration of the bubble into the droplet, yielding an oil encapsulated bubble whose geometry was similar to the one of the isolated droplet, whereas the terminal velocity of the aggregate formed increased by a factor of 95%, when compared to the original droplet. The injection of gas bubbles could, therefore, enhance the separation efficiency of oily wastewater.*

**Keywords:** *soft-particle, attachment, flotation, terminal velocity*

## 1. INTRODUCTION

A great challenge in fluid mechanics is the understanding and prediction of dispersed multiphase flows. Among these, flows with gas-liquid and gas-liquid-liquid interactions are frequent in industry, for instance, in applications from bubbly flow reactors up to wastewater treatment (Vélez-Cordero *et al.*, 2011). In this scenario, the removal of oil from wastewater has gained large attention over the years, in which the gas flotation technique has proven to be a viable option (Saththasivam *et al.*, 2016). This process consists in the injection of gas bubbles at the bottom of a flotation column, in which the interacting bubbles and oil droplets become attached to each other, yielding an aggregate with increased terminal velocity due to a larger density difference, promoting a more efficient separation (Moosai *et al.*, 2016). In the prediction of the interactions between individual soft-particles such as bubbles and droplets, the knowledge of parameters like the mean bubble diameter,  $d_b$ , and terminal velocity,  $\bar{u}_{\infty}$ , is required. With the considerations of low Reynolds number and spherical shapes, condition also designated creeping flow, this last parameter can be adequately modelled by the well-known Stokes approach. However, for larger bubbles, the increased buoyancy and terminal velocity often corresponds to a large Reynolds number as well as non-spherical shapes. Under this circumstances, the Stokes equation is not precise, with empirical relations normally being employed (Clift *et al.*, 1978).

In addition, the efficient application of flotation in wastewater treatment depends on the following steps: (1) collision between bubbles and droplets, (2) attachment and aggregate formation and (3) free rising aggregate (Saththasivam *et al.*, 2016). Therefore, the understanding of the attachment stage is crucial for the elucidation of the flotation technique. This phenomena is quite complex, depending both on the geometry of the dispersed phases as well as fluid interfacial and bulk properties, since the interaction zone between the bubble and droplet is under influence of interfacial phenomena like capillarity and Marangoni flows (Vélez-Cordero *et al.*, 2012). Regarding the geometry of the dispersed phases, recent studies indicated that smaller bubbles attach themselves more efficiently to oil droplets, due to the increased surface area and residence time. Also, the attachment of gas bubbles to an oil layer was studied by Lim *et al.* (2016), which observed the formation of a stable bubble-oil aggregate depending on the contact angle between the phases (Lim *et al.*, 2016).

However, the experimental evaluation of the interaction and attachment of gas bubbles to oil droplets under free gravitational motion is still not accounted for in literature. Under these circumstances, in addition to the interfacial phenomena, the hydrodynamics become also important, whose influence is far from being elucidated. Therefore, the present study aims to fill a large gap in this topic by experimentally analyzing the fundamental interactions and attachment

phenomena of gas bubbles to oil droplets under gravitational motion in a quiescent water medium. To this aim, the influence of the dispersed phases geometry in the attachment process and aggregate terminal velocity will be evaluated.

## 2. METHODOLOGY

Figure 1 illustrates the experimental set-up constructed in the Multiphase Flow Research Center (NUEM) in the Federal University of Technology - Paraná (UTFPR). Individual oil droplets and gas bubbles are generated in a distilled water medium employing capillary tubes placed at the bottom of a visualization chamber. Leaking was prevented by connection of a stainless steel sealed feedthrough (Spectite, USA). Pure nitrogen was used for the bubbles generation, whereas colored commercial corn oil was employed for the droplets, in which an adequate pressure is provided by a syringe pump (Teledyne Isco, USA). Both the gas and oil volumetric flowrates were controlled by needle valves. The interaction visualization chamber consists of an acrylic rectangular channel with dimensions of 20 x 10 x 20 cm<sup>3</sup>.

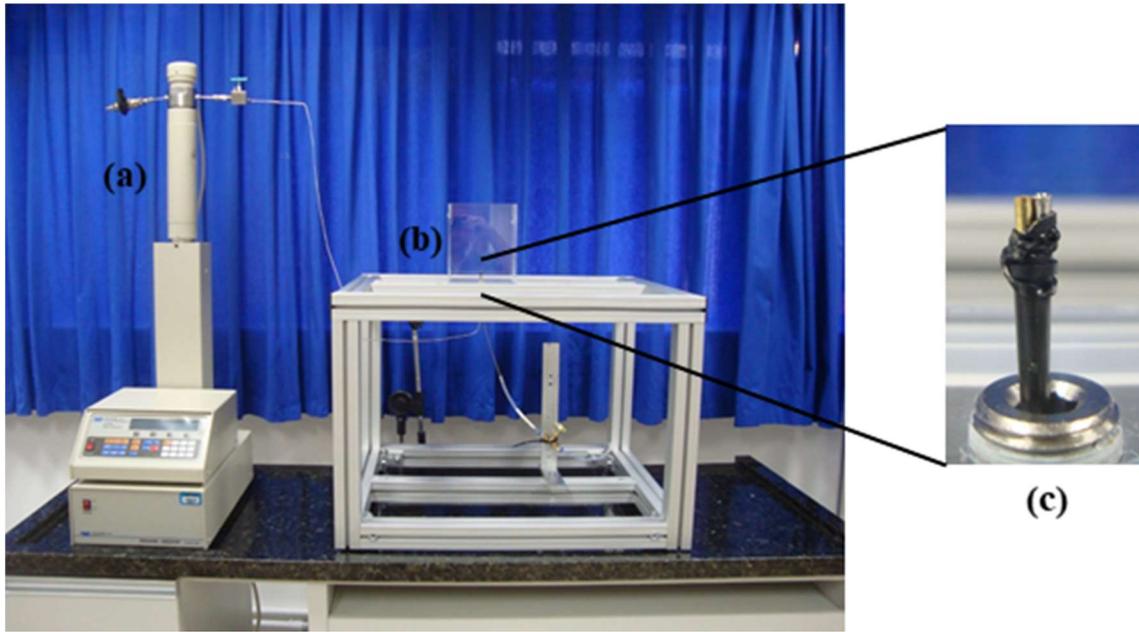


Figure 1. Experimental set-up employed in the attachment phenomena experiments. (a) syringe pump, (b) interaction visualization chamber and (c) capillary tubes arrangement.

Different capillary tubes were employed in the interaction assays, yielding gas bubbles from approximately 2.0 to 2.8 mm. In order to ensure individual and monodispersed bubbles, 300 mm length capillary tubes were employed, providing enough pressure drop so the bubble volume was independent from the volumetric flow rate (Vélez-Cordero and Zenit, 2011). Under these circumstances, the estimation of the inner diameter of the capillary tube,  $d_c$ , can be made considering a force balance between the buoyancy and surface tension forces, as exposed by Eq. (1).

$$d_c = \frac{d_b^3 \rho_w g}{6\sigma} \quad (1)$$

The interactions between the bubbles and droplets were analyzed using a high speed camera (NanoSense Mk III, IDT) with a acquisition rate of 500 frames per second. A specific Matlab code was developed by the authors for the image processing and parameters calculation, as briefly exposed in Fig. 2. The mean diameter was obtained considering an approximate ellipsoidal shape of the bubbles, according to Eq. (2) (Feng *et al.*, 2016).

$$d_b = (D^2 d)^{1/3} \quad (2)$$

where  $D$  and  $d$  are, respectively, the larger and shorter bubble diameter, as represented in Fig. 2. Another important geometrical parameter of the dispersed phases is the aspect ratio,  $\alpha$ , designated as the ratio between the larger and shorter diameter of the soft-particle, according to Eq. (3). It must be pointed out that for a spherical shape the aspect ratio is close to the unity.

$$\alpha = \frac{D}{d} \quad (3)$$

In addition to the aforementioned geometrical parameters, a physical characterization of bubbly flows is often carried out by employment of dimensionless parameters like the Reynolds,  $Re$ , and Eötvös,  $Eo$ , given respectively by Eqs. (4) and (5), where  $\rho_w$  and  $\Delta\rho$  are, respectively, the density of water and the density difference between the dispersed and continuous phases,  $\mu_w$  is the dynamic viscosity of water and  $\sigma$  represents the water surface tension. The Eötvös dimensionless parameter is of great importance since it compares the influence of inertial and interfacial forces, depending on the properties of the continuous and dispersed phases (Feng *et al.*, 2016).

$$Re = \frac{\rho_w \bar{u}_\infty d_b}{\mu_w} \quad (4)$$

$$Eo = \frac{\Delta\rho g d_b^2}{\sigma} \quad (5)$$

### 3. RESULTS

#### 3.1 Single bubble motion

A first stage of the experimental evaluation of the hydrodynamics and attachment phenomena between interacting gas bubbles and oil droplets is the single bubble motion characterization. This initial approach is of great importance, since the bubbles terminal velocity in the medium is from ten to one hundred times larger when compared with the one of the oil droplet (Moosai *et al.*, 2003), therefore, the bubbles tend to approximate the oil droplets as they rise, which makes clear that the bubble motion and trajectory greatly influences the collision, contact angle and attachment phenomena. Hence, geometrical and dimensionless parameters are calculated first for a single bubble in free gravitational motion. Figure 2 represents an arbitrary image processing of an individual bubble rising in distilled water. Through the binarization of the frame, the centroids position and the shape can be estimated, along with all other parameters. In the specific case shown, the mean diameter of the bubble was 3.11 mm, with an aspect ratio of 2.25 and a terminal velocity of 373.11 mm/s.

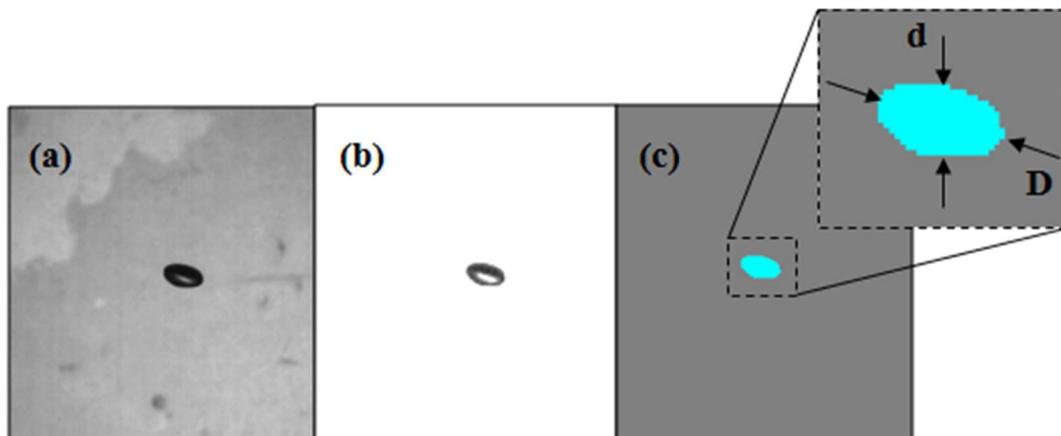


Figure 2. Individual bubble image processing. (a) original frame, (b) filtered frame and (c) binarized frame and geometrical parameters.

Further characterization of bubbly flows is made with the  $Re$ - $Eo$  plot, shown in Fig. 3. It can be seen that these two parameters are fundamental when estimating the shape of the bubbles, for instance, when  $Re < 1$ , the bubble tends to be spherical, with the Stokes equation being applicable. For higher values of  $Re$ , the bubble is spherical only for very low values of  $Eo$ , that is, when the surface tension forces are dominant over the inertial ones. For the present case, however, at room temperature, the Reynolds and Eötvös numbers are close to 700 and the unity, respectively, yielding a bubble of a wobbling shape.

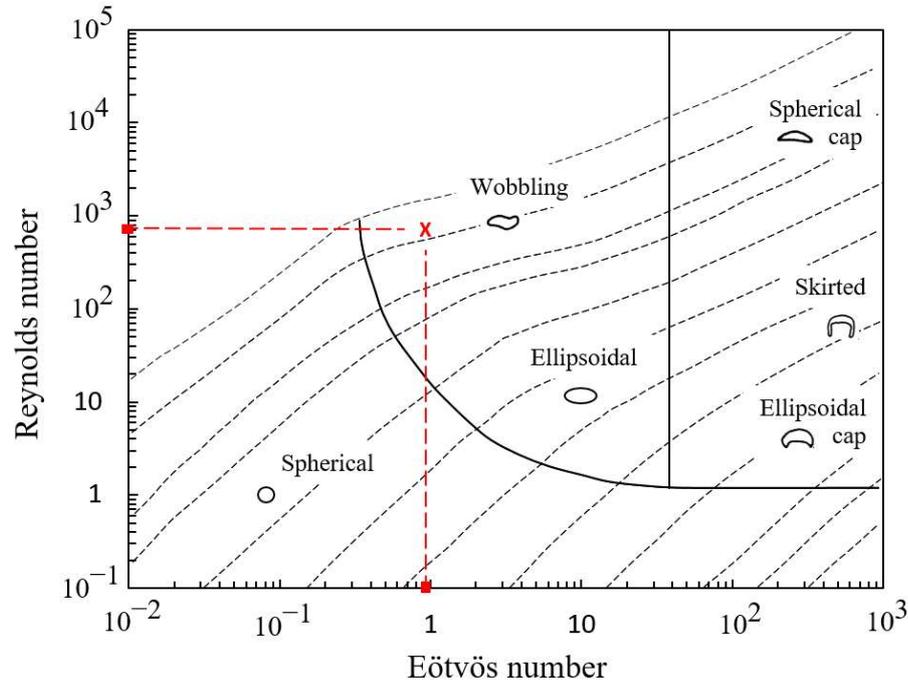


Figure 3. Shape regimes for bubbles and droplets in free gravitational motion through liquids (Adapted from Clift *et al.*, 1978). Notes in red represents the experimental single bubble experiment.

### 3.2 Individual bubble – droplet attachment

Once the basic knowledge of the bubbly flow is accomplished, interaction assays between individual gas bubbles and oil droplets can be carried out. Figure 4 exposes the images illustrating the attachment of a bubble to a droplet. Figure 4 (a) shows an isolated oil droplet with a diameter of 5.8 mm and aspect ratio of 1.25 just before detaching from the capillary tube. Once the gas bubble (with similar geometry to the previous single bubble experiment) expands from the capillary (Fig. 4 (b)) and contacts the droplets surface, an attachment phenomena is observed, in which the bubble penetrates the oil droplet and moves to its top, as shown in Fig. 4 (c – g). The increased density difference of the aggregate formed, due to the bubble penetration, induces the detachment from the capillary, as exposed in Fig. 4 (h – i) and, afterwards, free gravitational motion takes place, according to Fig. 4 (j – l).

Specific knowledge of the interfacial phenomena involved is required for the understanding of the attachment observed. As proposed by Hernández-Sánchez *et al.* (2015), the approach of the bubble to an oil droplet promotes the formation of a water lamella film, followed by drainage and rupture of this thin layer, due mainly to capillary and interface forces (Hernández-Sánchez *et al.*, 2015). After the rupture, an effective contact between the bubble and oil droplet takes place, leading to the attachment process.

This process evaluation can also be carried out through a spreading of liquid analysis, based on the physical chemistry properties of the phases involved. As the lamella film drains and finally ruptures, the bubble efficiently contacts the oil droplet and penetrate its surface, becoming encapsulated by the oil. In this process, one can state that a water-gas interface was substituted by two other interfaces (gas-oil and oil-water). In this scenario, a spreading coefficient,  $S_0$ , was defined, according to Eq. (6) (Moosai *et al.*, 2003).

$$S_0 = \sigma - \sigma_{go} - \sigma_{ow} \quad (6)$$

where  $\sigma_{go}$  and  $\sigma_{ow}$  represents, respectively, the interfacial tension of the gas-oil and oil-water interfaces. The spreading coefficient represents a balance between the interfacial forces in the system. A positive spreading coefficient indicates that the sum of the interfacial tension of the gas-oil and oil-water interfaces generated after the attachment is smaller than the water surface tension, related to the original isolated bubble. Under these circumstances, the attachment phenomena is energetic favorable, which was the case shown in this study. On the other hand, for a negative spreading coefficient, a bubble penetration in the droplet would not be energetic favorable, since the total interfacial energy would increase. Furthermore, the image processing showed no appreciable change in the geometry of the droplet after the penetration of the bubble, whereas the terminal velocity increased from 100.0 mm/s for the isolated oil droplet to 195.0 mm/s for the aggregate, representing an increase of 95 % in the terminal velocity and, therefore, a more efficient separation from the medium occurred.

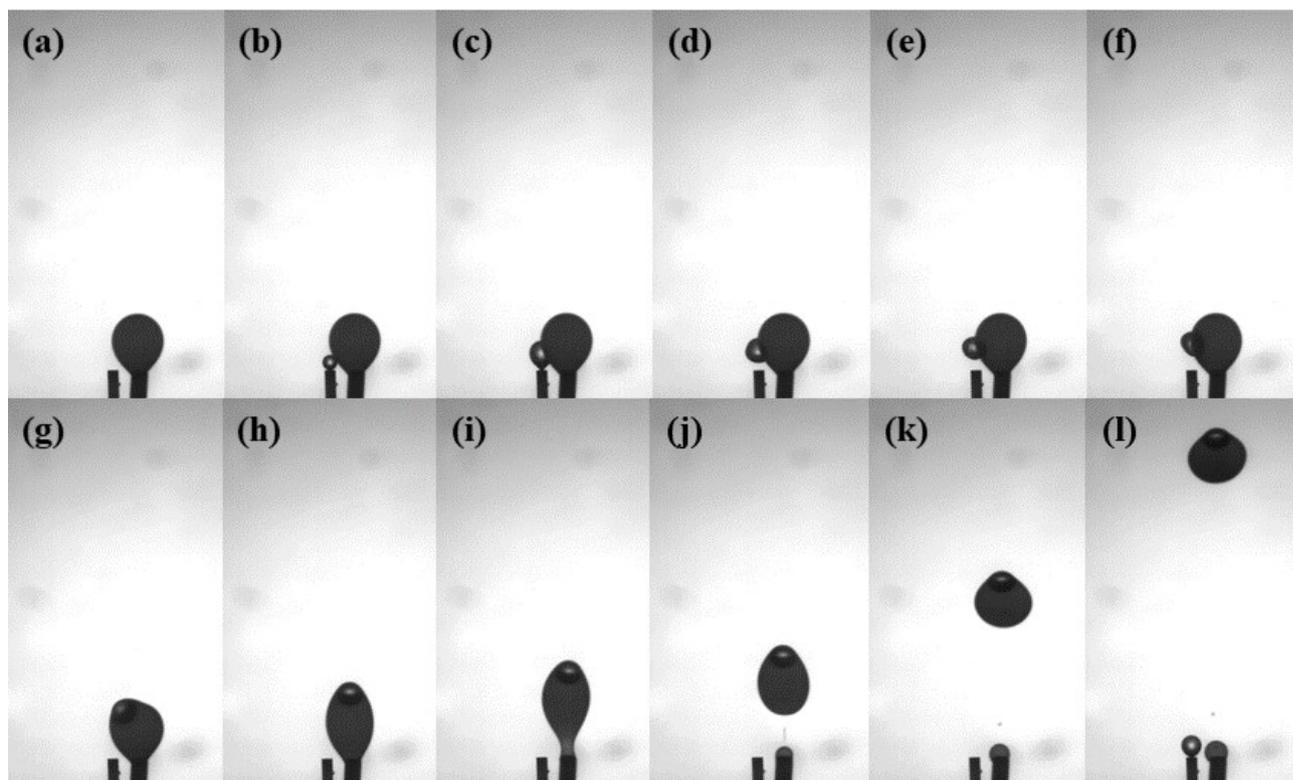


Figure 4. Individual gas bubble and oil droplet attachment phenomena. (a) isolated oil droplet, (b) gas bubble formation, (c) beginning of the interaction, (d-g) attachment and aggregate formation, (h-i) aggregate detachment and (j-l) aggregate free gravitational motion.

#### 4. CONCLUSIONS

The present work consisted of an experimental evaluation of the interaction and attachment of an individual gas bubble to an oil droplet, an applicable phenomena in flotation columns. High speed recordings was employed in all the assays of the present study. As a first stage, the free gravitational motion of a single bubble was characterized, where an individual bubble of 3.11 mm, aspect ratio of 2.25 and terminal velocity of 373.11 mm/s was generated. In addition to this preliminary stage, when a gas bubble and an oil droplet collide, an attachment process takes places, which is described as the penetration of the bubble into the droplet, becoming an oil encapsulated bubble, with the interfacial energy balance being favorable. After attachment, a stable aggregate is formed and the detachment from the capillary is induced, promoting a free gravitational rising motion with an increased terminal velocity and, therefore, separation efficiency.

#### 5. ACKNOWLEDGEMENTS

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