Hydrodynamic forces on floating particles

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Abstract

Detachment of particles from the bubble surface plays an important role in coarse particle flotation where hydrophobic and hydrophilic particles are separated from each other in an aerated aqueous pulp. The literature indicates that particle detachment from air bubbles depends strongly on the hydrodynamic forces acting on the particle. The objective of this study is to develop understanding about the role bubble deformation and shear flow has on particle detachment.

The stability of floating particles at an air-water interface has been historically developed using a thorough understanding of the supportive capillary forces to counterbalance the particle weight at equilibrium. As a particle undergoes detachment from an air-bubble, there are dynamic forces involved including bubble oscillation, centrifugal forces caused by bubble-particle rotation and shear flow past the bubble surface, which are not fully quantified in the literature and are difficult to measure individually. Bubble-particle vibration is well researched, with models to calculate detachment forces. Recent experimental work testing the centrifugal detachment theory has shown that the centrifugal force is not sufficient in itself to detach a particle from a rotating bubble. Emerging fluidisation technologies finding application in coarse particle flotation also have no models to predict detachment. In the literature, hydrodynamic corrections to Stokes drag force are only available for the case of floating particles translating across fluid-fluid interfaces in stagnant fluids. Corrections for fluid flow perturbations caused by a stationary, partially submerged particle in the normal and tangential directions to the interface are not readily available.

To test the hypothesis that shear flow is able to detach spherical particles (200 - 500 µm) from an air bubble, a new experimental technique was developed using a small-enclosed channel with fully developed laminar flow. Pure water flows past a partial bubble segment protruding from the channel wall at intermediate Reynolds numbers. Hydrophobic ballotini were attached to the air bubble and the water velocity increased, capturing particle movements using high-speed video microscopy. The results showed that detachment of spherical particles from a captive air bubble is possible, due to shear flow past the bubble surface. The results also showed that bubble deformation would occur when detaching forces were applied to the particle.

Deformation of the bubble interface during particle detachment was quantified using experimental results obtained with a modified sphere tensiometry apparatus detaching sub-millimetre spherical particles. A theoretical model was developed to describe the equilibrium shape of the bubble meniscus at any given particle position, based on minimising the free energy of the system. The
solution of the bubble profile was matched to the high-speed video calculating the contact angle and the total force balance as a function of the contact point of the bubble on the particle surface.

Measurements were undertaken to quantify the hydrodynamic drag forces exerted on a stationary spherical particle floating at the air-water interface under the influence of a shear and stagnation flow fields. This has been achieved using two new experimental techniques developed during this study. A shear flow field was generated in an open channel with pure water flowing past a partially submerged spherical silica particle. Using a carefully calibrated force cantilever attached to the particle, the drag force was calculated using Hooke’s law. A stagnation flow field was generated using a small vertical column where pure water flows upwards and past a partially submerged spherical silica particle suspended from a force balance directly measuring the exerted force on the particle. The results from the shear and stagnation flow fields are analysed, and a simple relationship as a function of the three-phase contact angle is developed for the drag force acting on a partially submerged particle in the tangential and normal directions to the air-water interface.

In summary, the aim of this work was to investigate the detachment of spherical particles from a bubble surface by shear flow. Measurement of the force required to detach a spherical particle from a deforming bubble in a quiescent fluid was undertaken and the results compared to an energy balance model. The drag force exerted on a stationary spherical particle floating at an air-water interface was measured separately using shear and stagnation flow fields. The Stokes drag force correction factor was reported as a function of the three-phase contact angle. To further understand the effects of bubble deformation and drag force on bubble-particle stability and detachment in practice, more research efforts are required to be undertaken. This should include, understanding the mechanism at the point of detachment where a trace bubble remains on the particle surface, and elucidating the differences in boundary slip conditions between a hydrophobic and hydrophilic particle surface. Finally, before a model predicting particle detachment in coarse particle flotation processes is fully developed, further study into particle shape effects on drag force should also be undertaken.
Declaration by author

This thesis is composed of my original work, and contains no material previously published or written by another person except where due reference has been made in the text. I have clearly stated the contribution by others to jointly-authored works that I have included in my thesis.

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Contributions by others to the thesis

Professor Anh V Nguyen contributed to the conception and design of the project, mathematical modelling, and writing in an advisory capacity.

Statement of parts of the thesis submitted to qualify for the award of another degree

None.

Research Involving Human or Animal Subjects

No animal or human subjects were involved in this research.
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An analysis of bubble deformation by a sphere relevant to the measurements of bubble-particle contact interaction and detachment forces

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Chapter 1

Introduction
1 Background

In the base metal processing industry, up until the last decade or so, coarse particle flotation has not been an area of research priority, compared to the practice of coarse particle flotation of phosphates that has occurred in Florida for over 40 years [1]. The driver to select a coarser grind in the comminution circuit is to reduce both the capital and operating costs for both Greenfield and Brownfield plants. Ore comminution requires the largest operational expenditure of mining operations consuming approximately 70% of the total energy requirements with up to 97% of this energy cost incurred during fine grinding of the ore [2].

Mechanical flotation cells are an effective technology for the recovery of mineral particles within the 20 to 150 µm size range [3]. Process plant data from various copper operations around the world illustrates the optimal size range as per Figure 1. The data shows that when the particle size reaches a certain threshold size the recovery drops off dramatically. The definition of coarse particle for sulphide ores in this context is for particles larger than about 150 µm.

![Figure 1. Copper Recovery from different concentrators](4)

Particle-bubble stability is of great importance in the flotation separation processes. The overall flotation process is usually divided into three sub-processes: collision, attachment and detachment. As will be discussed in Chapter 2 of this thesis, bubble-particle detachment is the rate limiting mechanism for successful coarse particle flotation. The literature indicates that particle detachment is primarily due to the hydrodynamic conditions occurring in a mechanical flotation cell. The widely used model predicting bubble-particle detachment probability is based on centrifugal motion [5]. The detachment theory was developed on the principle that centrifugal forces are exerted on attached particle when bubble-particle aggregates begin to rotate. The rotation occurs once the aggregates become trapped in turbulent vortices in the aqueous pulp. Based on this hydrodynamic knowledge, advancements in coarse particle flotation have recently been achieved. Researchers have
found that by utilising fluidisation principles, increased recovery is able to be achieved for coarse sized particles of up to 850 µm, due to the absence of turbulent vortices [6].

The centrifugal detachment model has recently been shown to be a primary detaching force for rotating bubble-particle aggregates, based on experimental results obtained [7]. However, analysis based on the well-established theory defining particle stability at an interface shows that the centrifugal model does not completely account for the total detaching force required. For coarse particle flotation technologies based on fluidisation principles, the centrifugal theory does not apply, due to the absence of rotating bubble-particle aggregates.

The research begins in Chapter 3 of this thesis where the shear flow past a bubble-particle aggregate is investigated. The experimental work showed that shear flow past a bubble surface was capable of detaching a spherical particle. This initial research identified two interesting points missing from the literature and warranting further investigation. The first interesting point arose while analysing the contact angles, as the contact angle was measured individually for each particle prior to detachment. The contact angle, which is discussed in more detail in Chapter 3, was calculated as the angle between the tangents at the point of intersection of two equations for a circle. The equations were obtained using captured images, fitting the bubble and particle profiles to the equation of a circle, respectively. This method relies on the assumption that there is no deformation of the interface, as the particle is much smaller than the bubble. However, when calculating the contact angles there were several cases when the equations describing the bubble and particle as circles failed to intersect. While these results were excluded from the investigation, it indicates the importance of a proper theoretical understanding of bubble deformation as a particle is dynamically detached. This problem is researched in further detail in Chapter 4, investigating the problem of bubble deformation as a particle undergoes detachment by developing an energy balance model to describe the process.

The second point of interest from Chapter 3 was that the results showed particle detachment required higher water flow rates with increasing hydrophobicity, as measured by contact angle. While, the maximum adhesive force was able to be calculated, based on a conventional force balance, no models exist to balance the well-established stability forces to the dynamic forces. This indicates the need to develop a new method to determine the drag force exerted on a stationary particle as a function of the contact angle and water velocity. This is important as some investigators attach separate significance to the parts of the total resistance arising from the integrated effect of the normal and tangential stresses, respectively [8]. Chapters 5 and 6 have been separated to individually measure and quantify the drag force on a stationary floating particle, for fluid flow in the tangential and normal directions to the air-water interface, respectively.
1.1 Research Objectives

This research project aims to develop insight into the role of bubble-particle deformation and the hydrodynamic resistance forces on the detachment mechanism related to coarse particles undergoing detachment from air bubbles. Specific objectives are as follows:

- Experimentally demonstrate that shear flow past a captive air bubble is capable of detaching a spherical particle from the surface and is a function of the contact angle and water velocity.
- Develop a new energy balance model for calculating the contact angle by deriving and solving a modified Young-Laplace Equation with a corresponding force balance during particle detachment from an air bubble.
- Experimentally investigate the shear flow drag force exerted on a particle attached to an air-water interface as a function of the three-phase contact angle and water velocity.
- Experimentally investigate the stagnation flow drag force exerted on a particle attached to an air-water interface as a function of the three-phase contact angle and water velocity.

2 Hypothesis

- The shear flow past a bubble is capable of moving/detaching a particle from the surface and is a function of the wetting properties.
- Deformation of the bubble surface occurs during detachment, due to contact angle hysteresis and this can be accurately described by an energy balance model.
- The hydrodynamic drag force exerted on a particle attached to an air-water interface can be correlated to the particles wetting properties as a correction to Stokes’ drag force equation, in the tangential and normal directions.

3 Structure of thesis

This thesis is organised into seven chapters. Chapter 1 introduces the project background, objectives and hypotheses. Chapter 2 is a literature review of the conventional force analysis, tenacity of attachment and hydrodynamic detachment forces. Chapter 3 gives results of experiments that demonstrate that shear flow past a captive air bubble is capable of detaching a spherical particle. Chapter 4 develops a new method to analyse the forces and contact angle hysteresis resulting from bubble deformation caused by the detachment of a spherical particle. Chapter 5 gives results of experiments that demonstrate the drag force on a stationary particle floating at an air-water interface in shear flow field is a function of the particle contact angle. Chapter 6 gives results of experiments that demonstrate the drag force on a stationary particle floating at an air-water interface in a stagnation
flow field is a function of the particle contact angle. Chapter 7 is the final chapter summarising the conclusions and provides recommendations for future work.
References


Chapter 2

Literature review on bubble-particle stability and detachment forces
Abstract

Detachment of particles from the bubble surface plays an important role in the efficiency of the flotation separation of hydrophobic particles from hydrophilic ones in an aerated pulp. The literature indicates that coarse particle flotation depends strongly on quiescent hydrodynamic conditions thereby minimising the probability of bubble-particle detachment. This review is concerned with understanding the current theories of force analysis and detachment modelling for particles of different shapes that describe the stability of the three-phase contact relevant to coarse particle flotation. The detachment of particles from an air-water interface has historically been based on an understanding to balance the supporting capillary forces in a system at equilibrium. The dynamic changes due to contact angle hysteresis as a spherical particle undergoes detachment from an air bubble has not been quantified in the literature. Recent experimental work testing the centrifugal detachment theory, has shown that developed centrifugal force in not alone sufficient to detach a particle from a rotating bubble and additional forces are required to be exerted. Emerging fluidisation technologies finding application in coarse particle flotation have no models to predict detachment. Hydrodynamic corrections to Stokes drag force for a floating particle translating across a stationary fluid-fluid interface are available in the literature. However, corrections for fluid flow perturbations exerted on a partially submerged stationary particle in the normal and tangential directions to the interface are not readily available in the literature, and this aspect is addressed in this work.

1 Introduction

Froth flotation has been widely used for over a century to separate valuable mineral particles from gangue particles [1]. Commercially the largest particles that can be floated are around 10 times smaller than that of the particles that can be theoretically floated. This was demonstrated by Gaudin et al. [2] who showed that for quiescent conditions sulphide minerals between 2 and 3 mm and 10.7 mm for bituminous coal were able to be supported at the interface. The recovery of minerals in commercial flotation cells is typically most successful in the 10-200 μm size range [3]. An important mechanism governing the size range of floatable particle is the motion of the solid particles and air bubbles and how they interact with each other in the pulp/slurry phase of a flotation cell. In order to optimise flotation performance, it is important to analyse the balance of forces acting on particles stabilising them at the interface. The capillary theory of flotation has historically been developed by studying the force when the system has reached a static equilibrium [4]. Only recently have researchers begun to study the dynamic bubble-particle detachment forces in isolation [5–9].

Floating particles are supported at the gas-liquid interface by capillary forces brought about by deformation of the interface. The profile of a deformed meniscus can be predicted using the well-
known Laplace–Young equation (YLE), which describes the relationship between hydrostatic pressure in the liquid and the pressure differential across the interface due to surface tension [10]. Solutions to the YLE have been developed with various analytical and numerical calculations published by researchers based on interface deformations caused by simple spheres and cylinders [4,11–13]. Using these solutions, researchers have established a quantitative link between the floating stability of a spherical particle and the surface hydrophobicity, quantified by the three-phase contact angle [14]. Other researchers, have developed mathematical models to predict the stability and detachment efficiencies for the bubble-particle aggregates [15–17]. Using the traditional force balance [14,18,19], the stability of a particle can be characterised by summation of the adhesive forces including capillary force, buoyancy and hydrostatic pressure. Despite these advances quantifying bubble-particle interactions, the physical principles underlying particle detachment from a free surface have not been fully explained. In the available theories, the dynamic nature of the detachment process has not been fully explored, and the contact angle is treated as a constant [7]. The hydrodynamic forces in a mechanical flotation cell are characterised by a single machine acceleration constant. However, contact angle may vary along the three-phase contact line and change with time during the detachment process due to pinning of the interface contact line arising from surface changes on the particle surface [20]. In addition, the energy dissipation rate in a stirred tank is distributed unevenly [21]. Further, bubble-particle detachment models for emerging technologies for coarse particle flotation based on fluidisation principles are not available in the literature.

The concept of coarse particle flotation in the mineral processing industries has received significant attention in recent years due to the potential for energy savings in the grinding process [22–27]. For the successful recovery of the coarse sized particles, it is well known that the particle detachment from air bubbles is the rate limiting mechanism. Reviewing the literature researching bubble-particle detachment in the pulp phase of a flotation cell found that there is no universal agreement upon a defining mechanism that accurately describes the detachment efficiency. However, particle detachment in the froth phase has been well researched [9,28] showing bubble coalescence is the primary detachment mechanism and while this is beneficial for cleaning the concentrate of gangue particles, this is not a focus of this review. For bubble-particle detachment in the pulp phase of a flotation cell, there are several proposed mechanisms. One proposed mechanism is that the sudden acceleration of bubble-particle aggregates creates bubble deformation resulting in particle detachment [29]. Alternatively, in his series of publications Schulze [18,30,31] developed a theoretical prediction for coarse particle detachment postulating that detachment was due to the centrifugal forces that are developed when a bubble-particle aggregate is caught within a turbulent eddy. This was experimentally tested by Goel & Jameson [21] who found that the detachment of particles follows a distribution of predicted values rather than a critical number. Additionally, several
different ‘kinetic energy’ models developed intuitively for particle detachment in the pulp phase have also been proposed [32–35], describing that bubble-particle detachment will occur if the particle collides with a bubble with a high level of kinetic energy.

Bubble-particle detachment forces have been measured using a bubble vibration technique developed by Holtham and Cheng [36,37] and further refined by Xu et al. [5]. The technique examined the detachment of coarse particles from oscillating bubbles as a function of particle hydrophobicity and shape, as well as medium viscosity. The results showed that the rate of movement of the three-phase contact line across the particle surface is reduced in liquids with a higher viscosity, resulting in a more stable bubble-particle aggregate. Atomic Force Microscopy (AFM) has been used to measure both the advancing and receding contact angles for individual colloidal particles. However, AFM has proved more useful for measuring interactions during the approach cycle rather than the detachment cycle [38,39].

Overall the literature does not describe a clear mechanism for bubble-particle detachment occurring in the pulp phase of a flotation cell. It indicates that it could arise as a result of either bubble deformation/oscillation, centrifugal forces or kinetic energy considerations does not provide a definitive conclusion as to which mechanism(s) is responsible for the detachment of coarse sized particles. To correctly evaluate the effect of the hydrodynamics of bubble particle interactions in flotation it is often necessary to return to the theoretical basis of hydrodynamics. Such effort requires understandably deeper studies of the hydrodynamic fundamentals and simplification of the problem by utilising knowledge of the fundamental theory [40–42]. The research and development of hydrodynamic theory into particle attachment and detachment mechanisms has historically resulted in the development of new experimental and theoretical knowledge of bubble-particle interactions. The water flow around bubble surfaces has been also studied in depth [42], with the aim to develop more accurate flotation efficiency models. This research focus on hydrodynamics has led to the development of new flotation technologies such as the Jameson cell [43], the Microcel™ [44], the Flash Flotation cell [45] and the HydroFloat Separator [46].

This literature review covers the current theories, which describe the stability of floating particles at an air-water interface. It introduces some of the important concepts in relation to the contact angle and its importance for successful flotation. The bubble-particle interactions that occur during the flotation process are also covered, with particular importance placed on the current understanding of the detachment process in relation to coarse particle flotation. The modified Stokes drag force equation is also discussed, regarding the implications for perturbed flow around air bubbles with attached particles on the interface.
2 Bubble-particle interactions in flotation

Flotation is used for the recovery of hydrophobic particles through their attachment to rising bubbles in an aerated slurry. In mineral processing froth flotation is used to separate valuable minerals from the gangue minerals present in the ore. Froth flotation is effective in separating particles, as depending on the mineral composition, the particle surfaces can be selectively rendered hydrophobic, by the addition of suitable flotation reagents. In the literature, interactions between bubbles and particles in a flotation cell is commonly separated into three sub-processes: particle-bubble collision, attachment and detachment. During each sub-processes there is different dominating forces, including hydrodynamic interactions, surface forces, and bubble–particle stability forces.

![Diagram showing bubble-particle interactions](image)

**Figure 1.** Schematic of the three key sub-processes occurring in the pulp phase of a mechanical flotation cell.

The breakdown into sub-processes, illustrated in Figure 1, was used by Derjaguin and Dukhin [47] who focused attention on the three distinct zones of bubble-particle capture. They considered that before a particle can adhere on the surface of an air bubble it undergoes influences from hydrodynamic, diffusiophoretic and surface forces for each zone it passes through as it approaches the bubble surface.

The overall probability of a hydrophobic particle being collected in the flotation concentrate by an air bubble in the pulp phase of a flotation cell can be described using

$$P = P_c P_a P_s$$

where $P$ is the probability of recovery, $P_c$ is the bubble-particle collision probability, $P_a$ is the attachment probability, and $P_s$ is the probability of bubble-particle stability. Collision and attachment probabilities were considered by Schuhmann [48] and the treatment was further expanded by Sutherland [49] who considered the effect of particle detachment.
2.1 **Bubble-particle collision**

The efficiency or probability of collision is defined as the ratio of the number of particles colliding with a bubble per unit time; to the number of particles approaching the bubble at a greater distance away with a cross sectional area equal to the projected area of the bubble [18]. The models predicting the collision efficiency are based on the premise that collision efficiency increases with increasing particle size for a given bubble size [35,50]. This means that the very fine particles tend to follow streamlines around the bubble circumference decreasing the probability of collision, while very coarse particles are much more likely to make contact with the bubble surface.

2.2 **Bubble-particle attachment**

For a bubble-particle aggregate to form, they first must come in close contact to each other, followed by the thinning and rupture of the liquid film forming a three-phase contact line [49]. The probability of bubble-particle attachment or adhesion is primarily determined by the achieved level of hydrophobicity on the target mineral surface. For hydrophobic particles to attach to a bubble surface there is a required induction time, first proposed by Sven-Nilsson [51]. The induction time, is the time taken for the liquid film to drain, rupture and form a stable three-phase contact line. For attachment to occur, the contact time; the sum of the impact and sliding times, must be longer than the required induction time for attachment. Hydrophobicity is not directly affected by the physical size of the particles. Rather the adsorption density of the collector should increase with particle size as larger particles have a smaller surface area. The particle size does not affect either the contact or induction times and is not a limiting mechanism for the recovery of coarse particles.

2.3 **Bubble-particle detachment**

Bubble-particle detachment occurs after the collision and subsequent attachment sub-process has occurred, and where the particle is unable to withstand the detaching forces created by the hydrodynamic drag or the turbulence in a flotation cell [52]. Another way to consider this is when the forces that hold particles at the surface are exceed by the detaching forces [14,53]. It has been shown by Nguyen [17] that the bubble particle detachment is characterized by nonlinear equations describing the gravity, capillarity, and buoyancy forces acting at the bubble-particle interface. Unlike the collision and attachment sub-process between bubbles and particles the detachment interaction is not satisfactorily quantified. The conventional approach has been to study the capillary theory and little consideration has been given to the dynamic forces involved as the particle undergoes detachment. The theoretical equations [35] for the collision and attachment efficiencies, suggest that flotation efficiency should continue to rise as the particle size increases, but the experimental evidence
suggests otherwise, indicating that the detachment sub-process becomes the rate-limiting mechanism for coarse particle flotation.

The main issues in relation to coarse particle flotation are firstly, to provide sufficient buoyancy to the particles by the air bubbles to lift the large particles up and into the froth. Secondly, are the detachment forces that come into play when large particles are rising in the liquid, in particular when they are near the impeller region in a mechanical cell. The last consideration is the forces that are exerted on coarse particles in the froth layer in the flotation cell that play an important role in cleaning the concentrate [22].

2.3.1 Mechanical cell detachment

The bubble-particle detachment sub-process has been shown by Deglon et al. [54] that the detachment mechanism can significantly influence the kinetics of flotation in a mechanical cell, due to the intensive turbulent agitation. The widely adopted cause for detachment found in the literature postulates that a centrifugal force is exerted on the attached particles when a bubble-particle aggregate is spun around in a turbulent eddy [14,18]. Another proposed mechanism of detachment is that the sudden acceleration of bubble-particle aggregates causes bubble deformation, resulting in particle detachment [29]. Several different ‘kinetic energy’ models for detachment in the pulp phase have also been proposed [32–35]. Initially, the kinetic energy model was described by Yoon & Mao [32] that bubble-particle detachment will occur if a particle collides with an bubble with a high level of kinetic energy after it momentarily attaches.

In a paper by Soto and Barbery [46] the authors state that conventional flotation cells operate with two contradictory goals. Firstly, the mechanical cell has to provide sufficient agitation to suspend and disperse the particles and air bubbles promoting bubble-particle collision. Secondly, to optimise recovery, by reducing particle detachment, a quiescent system is desired. As a result, coarse particle flotation is more difficult since increased agitation is required to maintain particles in suspension, while coarse particles are more likely to detach under increased turbulent conditions. This can be visualised in Figure 2.
2.3.2 Bubble size and clusters

The theoretical particle size limits to the flotation of coarse particles are imposed by the collective mass that can be lifted by one or more bubbles into the froth by the stable bubble-particle aggregates in a given turbulent flow field, and the disruptive energy released when the aggregate passed through the froth-liquid interface [30].

Bubble size plays an important role in floating mineral particles in either quiescent flow or turbulent conditions. Smaller bubbles are preferable for improving recovery of particles in a flotation cell with a constant gas rate and particle size distribution. The minimum bubble diameter [18] required to float a particle, can be estimated based on the principle where the mass of the bubble-particle aggregate must be less than the mass of liquid displaced. Therefore in a stagnant liquid:

$$d_{b, \text{min}} \geq d_p \left( \frac{\rho_p - \rho}{\delta} \right)^{1/3}$$  \hspace{1cm} (2)
where \( d_{b,\text{min}} \) is the minimum bubble diameter, \( d_p \) is the particle diameter, \( \rho_p \) is the solid density and \( \delta \) is the density of the liquid. According to Eq. (2), to lift 3 mm coal particles in the liquid, a 2.9 mm bubble is required. However, this size is somewhat larger than the size of bubbles generated in a typical flotation cell. Hinze [56] showed that for a given flow field there is a maximum stable bubble size, as the bubbles in a flotation cell respond to velocity gradients creating a velocity difference between two opposing apexes. This results in a pressure differential, causing the bubble to stretch and breakup when the surface tension force is exceeded by the stretching forces.

The formation of bubble clusters in flotation cells has been well known for some time since they were first noted by Gaudin [40] and Schulze [18]. Bubble clusters have also been observed in the new fluidisation based flotation cells [57,58]. Ata & Jameson [59] have proposed that cluster formation may be able to improve the flotation coarse particles, when a single large particle could be attached simultaneously to two or more bubbles, enhancing the buoyancy relative to single bubbles. Bubble clusters have been experimentally demonstrated to be created by the bridging of hydrophobic particles [59]. This means that the cohesive strength of the cluster is determined by the capillary force between bubble and particle. Bubble clusters have also extensively been studied by Chen [60] who showed the ability of their formation in process plant equipment. His research determined that the size of the clusters formed will range between 500 µm to 1 mm in industrial flotation cells, where the power input is typically in the range 0.5 to 5 kW/m³.

### 2.4 Coarse particle flotation

Coarse particle flotation has not been a central focus in base metals processing up until the last couple of decades. The reason for this is that there is a distinct trend to move towards a coarser grind size in comminution circuits to reduce the energy required to grind the ore [26]. A downside of coarse grinding is that lower flotation recoveries prevail with increased mineral losses in the coarser particle size ranges. The main issue to consider when operating at the upper flotation size range is that sufficient buoyancy is provided to lift the large sized particles from the pulp into the froth phase. Consideration of the hydrodynamic detaching forces that are exerted on the large particles rising in the fluid, especially when they are nearby to the impeller in a mechanical cell, is thus very important. Split conditioning to improve the recovery of coarse particles [61] is where the coarse and fine particles of a flotation feed are separated, separately dosed with collector, and then recombined for flotation. Another approach for treating coarse composites in beneficiation plants is the use of technologies such as the Flash flotation cell [25]. Commercial adoption of fluidisation technologies to reduce the turbulence normally encountered in conventional flotation cells has also been adopted [62].
The primary grind size of the ore has a major impact in the design of a flotation circuit. Flotation response depends on the level of liberation of the minerals in the ore [63]. Typically, the valuable mineral to be separated by flotation is initially embedded in a host rock from which it needs to be liberated by crushing and grinding. The liberation characteristics of particles play an important role in the treatment requirements for flotation and this has been an area of considerable research input and is covered extensively [64–71].

It is well established that the recovery of particle by flotation is most successful in the 10-200 µm size range [35,61,72]. The recovery of fine particles by flotation is usually low, due to the significant decrease in the collision rate and encounter efficiency as the particle size decreases. This agrees with the hydrodynamic theory, which predicts that the collision rate and the encounter efficiency are proportional to the particle size [21,40,49,73,74]. Some typical size recovery data for several sulphide minerals is given in Figure 3 (after Jowett [75]). However, for coarse particles that are larger than 200 µm, another set of problems is presented.

![Figure 3](image.png)

**Figure 3.** Size by size recovery for several common sulphide minerals, obtained from batch flotation tests after 1 minute [75].

A key question in terms of coarse particle floatation is the extent to which mineral liberation influences the performance. Wang and Fornasiero [69] used synthetically generated composite particles to study the flotation behaviour of particles between 75 µm to 600 µm in mechanically agitated flotation cell. The particles were classified as either simple or complex composite particles. They found that the flotation recovery was reduced for increasing size fractions and a reduction in the valuable mineral content. Interestingly, it was observed that for the particles with complex locking textures the flotation recoveries were found to be higher than for particles with simple locking textures for the same size interval. It was postulated that the complex textures allowed for multiple
hydrophobic areas available for bubble-particle contact. Recently Jameson [76] analysed data obtained by Welsby et al. [77] and concluded that “poor recoveries of coarse particles has nothing to do with poor liberation… the decline in recovery of coarse particles is related to the hydrodynamic conditions in the flotation cell.” This statement was based on the premise that the peak in the flotation rate constant as a function of particle size is found for fully liberated particles. This is best illustrated by Figure 4.

![Figure 4. Rate constant data from Welsby et al. [77] plotted against mean particle size (after Jameson [76]).](image)

It is postulated that the primary reason for poor recovery of the coarse sized particles can be traced to the high levels of turbulence present in a mechanical flotation cell. Jameson et al. [22] showed, based on the centrifugal detaching force, that the maximum size of a particle that can be floated in a turbulent mechanical flotation cell can be estimated by:

\[ d_{p\text{ max}} = 1.53 \sqrt[6/5]{\frac{\sigma^{6/5} (1 - \cos \theta)}{\Delta \rho \delta^{2/5} \varepsilon^{2/5}}} \]  

where, \( d_{p\text{ max}} \) is the maximum diameter of the particle that can be floated, \( \varepsilon \) is the power per unit mass based on the mass of liquid in the volume swept by the impeller, \( \delta \) is the density of water and \( \theta \) is the three-phase contact angle.

Recovery of the valuable minerals depends on two factors; the bubble-particle interfacial chemistry and the hydrodynamic conditions present in the flotation cell [78]. Experimental results shown in Figure 3 reveal that when the particles reach a certain size the recovery drops off dramatically. As the probability of collision is increasing this leads to the conclusion that the mechanism for particle-bubble detachment due to the hydrodynamic conditions in the flotation cell is the contributing cause for the decreased recovery of coarse particles.
The premise that the hydrodynamic conditions are the only factor that determines the maximum floatable particle size is probably not entirely correct. As Welsby et al. [77] reported that for the coarse low grade particles that did not float, they were unable to determine if this occurred due to the particles being unable to attach to the bubbles, or early detachment in the pulp or across the froth phase. Recovery of coarse particles in a mechanical flotation cell suffers the most because the bubble-particle aggregate forces are not strong enough to prevent the dynamic forces present from detaching the coarse sized particles.

### 2.5 Coarse particle flotation applications

The technology for the Flash Flotation cell, illustrated in Figure 5, has been around for some time, with the first documented unit installed in Finland in 1982 [45]. This technology has a special application in the grinding circuit to produce a concentrate whilst preventing over-grinding of the liberated values in the ore. The purpose of the flash flotation device is indeed to recover the very well liberated material within the floatable size range, or the fast floating material.

![Flash flotation cell](image)

**Figure 5.** Flash flotation cell [25]

Flash Flotation cells are designed to take a coarse feed material, typically from the cyclone underflow, after primary grinding with the feed often containing small rocks. The inlet/outlet of these cells is designed specifically for the coarse material to bypass the cell to the tailings stream, this allowing the finer particles to be drawn into the impeller region. The impeller (rotor/stator) set up is not dissimilar to a conventional tank cell. Work conducted by Newcombe et al. [79] found that coarse particles around 850 µm in size are collected by bubbles in the region directly above the impeller, but as the bubble-particle aggregates proceed to rise through the cell and approach the overflow launder the coarser sized particles begin to detach from the bubbles.

The idea of fluidising particles for flotation applications was patented by Barbery et al. [62]. The HydroFloat separator was later developed and can be described as an air-assisted, fluidised-bed
The cell operates based on the theory of density separation and using the fundamentals of flotation. The unit is constructed with an upper separation chamber and a lower dewatering cone. Air is injected into the elutriation water which then passes through spargers distributing the air/water across the area of the separation chamber, fluidising the particles. Due to the high water flow to create fluidisation, the HydroFloat cell operates with minimal froth layer (see Figure 6).

![Figure 6](image_url)

**Figure 6.** A schematic diagram of the HydroFloat separator [81]

During operation the feed particles fall against a rising current of water, hydrophobic particles attach themselves to the bubbles reducing the settling velocity. The particles with increased buoyancy rise through the particle bed to be collected as concentrate, while the hydrophilic particles sink through the fluidised-bed and report as high density tailings (at about 75% solids) [57]. The HydroFloat separator was originally designed for the flotation of industrial minerals in particular coarse phosphate flotation, for example the Mosaic’s South Fort Meade operations. Over the past 15 years it has found commercial application in the flotation of feldspar, potash and diamonds with some proof of concept tests conducted using sulphide minerals and coal [82]. Results for the flotation of coarse sulphide particles, in the range of 250–1180 μm, indicated that for the very coarse particles (+425 μm) the HydroFloat separator has a higher recovery compared to a conventional mechanical cell. The higher recovery observed was attributed to the absence of both turbulence and froth phase, both of which are generally considered detrimental to coarse particle flotation.

In addition to developing the Jameson cell [43], Jameson has also recently developed a fluidised bed flotation cell, illustrated in Figure 7. In this design a portion of the slurry is recycled to create the fluidised bed suspending a substantial fraction of the particles so that the coarse particles remain in the bed. The recycled water is aerated dispersing the air into fine bubbles. The purpose of this design is to have a device that can capture particle over a wide size range.
Figure 7. Sketch of a fluidised bed NovaCell flotation cell [83]

Jameson [84] carried out a set of flotation tests on a prototype device, comparing the results to work performed by Jowett [75] using a conventional mechanical flotation cell. For galena Jowett was able to achieve high recoveries over the range of 8 µm to 150 µm. For the NovaCell the range of galena with high recoveries was between 200 µm and 850 µm.

2.6 Bubble-particle detachment studies

Theoretical and experimental studies conducted by Schulze [18,30] showed that the upper particle size limit for flotation can be calculated by balancing forces acting on a bubble-particle aggregate based on the centrifugal detachment theory. His work showed that turbulent conditions, typically found in mechanical flotation cells, drastically reduce the upper size limit of floatable minerals. Schulze [31] proposed a dimensionless number, Eq. (5), analogous to the Bond Number, to predict the probability of particle detachment. The Bond Number is defined as the ratio of the detaching forces to the attaching or adhesive forces. When a Bond Number (Bo) is less than 1 the particles will float and when the Bond Number is greater than 1 the particles do not float. The Bond number is given by:

\[
Bo = \frac{F_g - F_b + F_p + F_a}{F_c}
\]  

(4)

\[
Bo = \frac{d_p^2 (\Delta \rho g + b_m) - d_v \sigma \cos^2 (\theta/2)}{6 \sigma \cos^2 (\theta/2)}
\]  

(5)

\[
Bo_m = 3.75 \frac{d_p^2 \rho (\epsilon^{2/3} d_h^{1/3})}{6 \sigma \sin^2 (\theta/2)}
\]  

(6)
where $d_p$ is the particle diameter, $d_b$ is the bubble diameter, $\varepsilon$ is the energy dissipation rate, and $b_m$ is the machine acceleration constant.

An experimental study to validate the centrifugal bubble-particle detachment theory was carried out by Goel [85] in a specially designed agitated tank, shown in Figure 8. The experimental tank uses an impeller with a well-defined power input, operated with no froth phase, and was able to separately collect the detached particles from the feed and recovered overflow.

![Experimental setup to assess particle detachments from bubbles in an agitated tank, specially designed to capture the detached particles [21].](image)

**Figure 8.** Experimental setup to assess particle detachments from bubbles in an agitated tank, specially designed to capture the detached particles [21].

Based on this research a modified Bond number was proposed by Goel [21], Eq. (6). The results showed that the detachment of particles follows a distribution of predicted values rather than a critical number. However, as the energy dissipation rate in the stirred tank is distributed unevenly, how the particles detached from the bubbles due to the turbulence is not clear.

The detachment of coarse particles from oscillating bubbles as a function of particle hydrophobicity, shape, and liquid viscosity has been experimentally studied by Holtham and Cheng [36,37] and later by Xu et al. [5,86,87]. The detachment experiments used various levels of hydrophobicity and sizes of both quartz particles and spherical glass particles. A schematic of the experimental setup used is included in Figure 9.
Figure 9. A schematic diagram of the apparatus used to study the detachment of particles from bubbles [86].

The detachment force of the bubble-particle aggregate can be calculated by the summation of the maximum vibrational force and the buoyancy force. The detachment force for the particles was calculated based on the maximum vibration amplitude when the test particle detaches from the oscillating bubble. The results found that, with an increase in both the contact angle of particles and viscosity of the suspending medium, the required force to detach the particles increased [87]. It was hypothesised, that the magnitude of the detachment force is determined by the dynamic contact angle, which is governed by the rate of movement of the three-phase contact line across the particle surface. Xu et al. [87] observed that larger detachment forces were obtained for irregular quartz particles compared to that of the spherical glass beads for the similar particle sizes and contact angles.

An experimental study into bubble-particle detachment in a turbulent vortex to test the centrifugal theory of detachment has been recently undertaken [88]. The experimental set-up used is illustrated in Figure 10. A rotating flow field develops within a small wall cavity as water flows through a channel. A bubble that was pre-loaded with one or more particles was introduced into the cavity and by using a high-speed video camera the motion of the bubble-particle aggregate was recorded.

Figure 10. Schematic diagram of water channel system to develop a vortex created by turbulent flow [88].
Analysis of the video showed that the induced rotational motion of the bubble particle aggregate was able to detach a particle from an air bubble. An analysis of the force balance, based on using the modified Bond Number, showed that detachment occurred at a value less than 1. The experimental results obtained are plotted in Figure 11. The low values of the Bond numbers obtained, indicates that the calculated centrifugal force does not wholly account for the detachment of the particles from a rotating bubble, with some additional unidentified detaching force is also involved in the detachment process.

**Figure 11.** Calculated Bond Number of the particle at the point of detachment from a rotating bubble [88].

### 3 Stability of spherical particles at an interface

#### 3.1 Classical force balance for a floating particle

The capillary theory of flotation was developed for spherical particles attached to a planar gas-liquid interface as illustrated in Figure 12. The modelling of the forces holding a particle floating at the interface have been well established [4,35,89]. The balance of forces describing stability of a spherical particle can be developed mathematically based on an intuitive force balance using the geometry defined in Figure 12. For a floating particle in a quiescent system, there are three main adhesive forces acting in the upwards direction, supporting the particle at the interface, counterbalanced by the particle weight.
**Figure 12.** Schematic of a spherical particle attached to a much larger air bubble [35]. Where $\theta$ is the particle contact angle, $\alpha$ is the central polar angle, $\beta$ is the angle of inclination of the gas liquid meniscus at the three-phase contact, and $H$ is the height of the deformed meniscus, $R_p$ is the particle radius, and $\sigma$ is the gas-liquid surface tension.

The most important force stabilising the particle at the interface is the surface tension force [17]. The surface tension acts along the tangent at the three-phase contact point of the gas liquid interface, pulling the particle upwards vertically into the gas phase. The magnitude of this force is equal to the product of the gas-liquid surface tension and the length of the three-phase contact line. The surface tension force can be separated into horizontal and vertical components. The horizontal component of the forces can be negated, due to the rotational symmetry around the vertical axis. The vertical component of the capillary force ($F_c$) can be described by:

$$F_c = 2\pi R_p \sigma \sin \alpha \sin(\theta - \alpha) \tag{7}$$

The buoyancy force ($F_b$) is the second supporting force for the particle at the interface and is defined by Archimedes' principle. This force is dependent on the volume of liquid displaced by the fraction of the particle immersed in the fluid, giving:

$$F_b = \frac{\pi R_p^3 \delta g}{3} \left(2 + 3\cos \alpha - \cos^3 \alpha \right) \tag{8}$$

where $\delta$ is the density of liquid and $g$ is the acceleration of gravity constant.

The hydrostatic pressure ($F_p$) arising from the height of the deformed gas-liquid interface, $H$, acts over the area enclosed by the three-phase contact line above the contact area and given by the [17]:

$$F_p = \pi R_p^2 \sin^2 \alpha \ \delta g H \tag{9}$$

The gravitational force ($F_g$) exerted due to the weight of the particle, tending to pull the particle into the liquid phase is given by:
\[ F_g = \frac{4\pi}{3} R_p^3 \rho g \]  

(10)

where \( \rho_p \) is the particle density.

For a quiescent system, the balance of the forces described, at equilibrium must equal zero i.e. \( F_c + F_b + F_p + F_g = 0 \). These forces determines the stability of a floating particle and rewriting Eqs. (7) - (10) we obtain the following useful force balance equation:

\[
2\pi R_p \sigma \sin \alpha \sin(\theta - \alpha) - \frac{\pi R_p^3 \delta g}{3} \left(2 - 3 \cos \alpha + \cos^3 \alpha \right) + \pi R_p^2 H \delta g \sin^2 \alpha = \frac{4\pi R_p^3 g \Delta \rho}{3}
\]

(11)

As has been shown in the literature [90,91] undertaking a force balance for complex capillary systems can lead to errors. Another more rigorous approach for describing the stability of bubble-particle aggregates can be performed by an analysis of the system energy. Here the conditions for mechanical equilibrium can be determined through performing a minimisation of the free energy for a given capillary system, using the calculus of variations method [92]. This method has been used by Nguyen [35] to carry out the minimisation of free energy for a particle floating at an planar interface. Analysis of the capillary system, illustrated in Figure 12, leads to the confirmation that Eq. (11) is correctly balanced for the forces acting on a floating spherical particle in a quiescent system.

3.2 Force balance for a particle attached to a bubble

Figure 13, shows an illustration for a particle attached to a bubble surface with a slightly deformed meniscus caused by the particle weight. The forces between a bubble and particle in this circumstance can be analysed using the same force balance principles as discussed previously, by the inclusion of capillary pressure into Eq. (9). This results in the equation for the pressure force becoming:

\[ F_p = \pi R_p^2 \sin^2 \alpha \left(\frac{2\sigma}{b} - \delta g H \right) \]

(12)

where \( b \) is the bubble radius at the apex.
Modification of the force balance for a dynamic system viz. the pulp phase of a mechanical flotation cell, means additional detaching forces can be represented by a particle acceleration constant for a given system [31]. The equation for representing this additional detaching force in a dynamic system can be represented by:

$$F_a = \frac{4\pi}{3} R_p^3 \Delta \rho b_m$$

(13)

where the density difference between the particle and the liquid and is defined by the formula $\Delta \rho = (\rho_p - \delta)$. The machine acceleration constant describing the bubble-particle aggregate acceleration can be determined using:

$$b_m = 1.9 \frac{\varepsilon^{2/3}}{d_B^{1/3}}$$

(14)

where $\varepsilon$ is the energy dissipation rate and $d_B$ is the bubble diameter. Additional revisions to Eq. (14) have been provided by others to improve the estimate for the machine acceleration constant [21,93]. For a comprehensive review of the available calculation methods see the work of Wang et al. [94]. The machine acceleration constant is applicable for modelling detachment in the pulp phase of a conventional mechanical flotation cell. In fluidised bed flotation, where bubble-particle aggregates are supported by a rising flow of fluid, Eq. (13) is not a valid theory to be applied due to the absence of turbulent vortices [95].

### 3.3 Tenacity of particle attachment

The stability of particle attachment at the gas-liquid interface is a strong function of the hydrophobic nature of the particle surface, the higher the level of hydrophobicity, the more stably it remains attached to the interface. Analysing a summation of the adhesive forces, Eqs. (7) - (9),
described by Eq. (15), shows the adhesive force to be a strong function of the polar position of the three-phase contact line (see Figure 14).

\[
F_{\text{adhesive}} = 2\pi R_p \sigma \sin \alpha \sin (\theta - \alpha) - \frac{\pi R_p^3 \delta g}{3} \left( 2 - 3 \cos \alpha + \cos^3 \alpha \right) + \pi R_p^2 H \delta g \sin^2 \alpha
\]  

(15)

The adhesive force balance described by Eq. (15) can be found to have a maximum force, illustrated in Figure 14, when the value of \( \alpha \) is equal to half the contact angle. The particle is detached from the interface only if the detaching forces exceeds this maximum value, which is called the tenacity of attachment [15].

![Figure 14](image.png)

**Figure 14.** Plot of the adhesive force, described by Eq. (15), supporting a spherical particle at an air-water interface as a function of the central polar angle [17].

By studying the results of numerical solutions to the Young-Laplace equation, describing the deformation of the interface [17,96], simplification of the adhesive force balance to calculate the maximum supporting force, yields:

\[
T = \pi R_p \sigma \left( 1 - \cos \theta \right) \left\{ 1 + 0.016 \frac{R_p}{L} \right\}
\]  

(16)

where \( T \) is the tenacity of attachment and \( L \) is the capillary length defined by \( L = \sqrt{\frac{\sigma}{\delta g}} \).

### 3.4 The Young-Laplace Equation

The shape of the deformed gas-liquid meniscus caused by the presence of an attached particle is formed by the equilibrium balancing both sides of the interface by the surface tension and hydrostatic pressure. The curvature of the surface can be described using the Young-Laplace equation, which relates the pressure difference, \( \Delta P \), across the interface, and the surface tension by:
where $\kappa$ is the mean curvature of the gas-liquid interface.

The hydrostatic pressure at any point on the meniscus surface can be determined by using the well-known equation:

$$\Delta P = \delta gh$$  \hspace{1cm} (18)

where $h$ is the vertical coordinate of the meniscus. In the case of a floating spherical particle where the system is rotationally symmetrical, as shown in Figure 12, the expression of the Young-Laplace equation can be transformed using terms of the first and second derivatives of the meniscus depth $h$, with respect to the radial coordinate $r$, resulting in: [35]

$$\sigma \left\{ \frac{d^2 h}{dr^2} \left[ 1 + \left( \frac{dh}{dr} \right)^2 \right]^{3/2} + \frac{dh}{dr} \left[ 1 + \left( \frac{dh}{dr} \right)^2 \right]^{1/2} \right\} = \Delta \rho gh$$  \hspace{1cm} (19)

Eq. (19) can be numerically solved to obtain the shape of the interface profile, achieved by converting the differential equation into a set of solvable parametric equations by using the arc length, $s$, of the meniscus [13], yielding:

$$\frac{dx}{ds} = \cos \phi$$
$$\frac{dy}{ds} = \sin \phi$$
$$\frac{d\phi}{ds} = \frac{y - \sin \phi}{x}$$  \hspace{1cm} (20)

where $\phi$ is the angular inclination of local bubble meniscus to the $r$-axis, i.e. $dh/dr = \tan \phi$, and depending on the boundary conditions can be used to calculate the contact angle, $\theta$, if $\alpha$ is also known. The parameters $x = r/L$ and $y = h/L$ are dimensionless variables to simplify the integration. The set of the parametric equations can be integrated using a fourth-order Runge-Kutta scheme specifying boundary conditions at the three-phase contact line. The numerical solution to the Young-Laplace equation can also be used to provide additional information for the volume of displaced air, $V_b$, and the surface area, $A_b$, defined as:

$$V_b = \pi \int_0^{s_{\text{max}}} r^2 \sin \phi \, ds = \pi R_p^3 \frac{2 - 3 \cos \alpha + \cos^3 \alpha}{3}$$  \hspace{1cm} (21)

$$A_b = 2\pi \int_0^{s_{\text{max}}} r \, ds$$  \hspace{1cm} (22)
Results for Eqs. (21) and (22) can be obtained simultaneously together with Eqs. (20) using the fourth-order Runge-Kutta scheme.

4 Contact angle and implications for floating particles

An important measure of the hydrophobicity of a solid surface is the three-phase contact angle, $\theta$, illustrated in Figure 15. The contact angle, as described by Young [97] can be determined as a mechanical equilibrium balance of the three interfacial tensions. The balance of surface forces can be written as:

$$\sigma_{sg} = \sigma_{sl} + \sigma_{lg} \cos \theta_i$$  \hspace{1cm} (23)

where $\theta_i$ is the ideal contact angle, $\sigma_{sg}$, $\sigma_{sl}$ and $\sigma_{lg}$ are the surface tensions between the solid-gas, solid-liquid and liquid-gas phases. However, the apparent contact angle is sensitive to a number of environmental factors, which makes obtaining a meaningful and reproducible contact angle values for mineral particles encountered in flotation, a real challenge. The value of the measured contact angle is affected by many variables; including; surface roughness, heterogeneity and particle shape.

![Diagram of contact angle](image.png)

**Figure 15.** Contact angle, $\theta$, between bubble and mineral surface in water, note the angle is measured through the liquid phase.

The contact angle and its importance in flotation has been studied for well over a century [1,98,99]. The contact angle, as shown in Figure 15, is measured at the three-phase contact line of an air bubble attached to a solid surface in an aqueous solution. For sulphide minerals to successfully float in a mechanical flotation cell, the contact angle required for flotation is around 60-80° [100]. For coarse particle flotation based on fluidisation principles the critical contact angle for flotation for a sulphide mineral has been shown to be reduced to between 40-60° [100]. As the contact angle reduces and approaches zero, no flotation will occur as the surface becomes fully hydrophilic.

The contact angle, while indicating the level of hydrophobicity of the minerals is not a thermodynamic parameter, it is, nevertheless, an important parameter for indicating successful
flotation. It has been observed by Blake and Ralston [101] that for different mineral particle sizes there is a varying critical contact angle required for flotation to occur. Crawford & Ralston [102] studied the flotation of methylated quartz particles using a modified Hallimond tube, the results are plotted in Figure 16. They observed the formation of a clear flotation domain, where flotation occurs based on reaching critical contact angle.

![Figure 16](image)

**Figure 16.** Flotation threshold values of advancing contact angle as a function of particle size for methylated quartz particles using a modified Hallimond tube [102].

It should be noted that the flotation domain, observed in Figure 16, is unique to the specific system and is not universal being strongly dependent on the hydrodynamic conditions of the apparatus. Flotation recovery, as previously mentioned is dependent on a wide number of parameters broadly including; engineering, physical, and chemical ones.

The surfaces of mineral particles typically encountered in industrial flotation cells are rough and chemically heterogeneous. The sharp corners and edges of the mineral particles due to the crushing and grinding have a significant effect on the contact angle measurement. The Wenzel equation [103] was developed to describe the changes in surface roughness causing fluids to have more pronounced hydrophobic or hydrophilic behaviour. This equation is applicable when the roughness scale allows the liquid to completely penetrate into the grooves on the solid surface.

\[
\cos \theta_w = \sum r_i \cos \theta_i
\]

where \( \theta_w \) is the Wenzel contact angle and \( r_i \) is the ratio of the true area to the apparent area.

The apparent contact angle will change again, due to the heterogeneous nature of the mineral particle surfaces resulting from the degree of liberation of the mineral grains. To describe a heterogeneous surface with different levels of hydrophobicity, the apparent contact angle can related by the Cassie equation to the ideal contact angle for each surface [104].
\[ \cos \theta_c = \sum f_i \cos \theta_i \]  

where \( \theta_c \) Cassie contact angle and \( f_i \) The fractional area with a contact angle \( \theta_i \).

### 4.1 Contact angle hysteresis

The validity of the Young equation, Eq. (23), stipulates that the surface of the solid must be smooth, flat, homogenous, inert, insoluble, nonreactive and non-deformable. However, in practice almost all surfaces are not ideal, and the contact angle measured is the apparent contact angle. When the three-phase contact line is under motion, the contact angle formed is referred to as the “dynamic” contact angle.

![Figure 17](image)

**Figure 17.** Contact angle on an inclined plate a) advancing contact angle and b) receding contact angle

The movement of the contact line across a surface are referred to as the advancing contact angle, \( \theta_A \), and receding contact angle, \( \theta_R \), illustrated in Figure 17. The magnitude of the measured dynamic contact angles will be different depending on the rate of motion of the three-phase contact line [39]. The difference between the advancing (largest) and the receding (smallest) contact angle is referred to as contact angle hysteresis and is defined by:

\[ \Delta \theta = \theta_A - \theta_R \]  

The importance of contact angle hysteresis in flotation has been well known for some time [105]. In most cases, it is unavoidable due to the nature of real surfaces. Given the dynamic nature of flotation, the study of contact angle hysteresis is important in understanding the bubble-particle detachment process in flotation. Recent studies [20,106,107] have considered the case where the contact line has been pinned at a sharp edge on a particle surface. In a study applying AFM conducted by Ally et al. [20] using particles with circular cuts, their work clearly showed that the contact angle at the edge could take a range of values within the range defined by the Gibbs inequality condition [106,108].

Recently, an excellent review [109] of the available methods to measure contact angles has been published. Details of the various experimental procedures have also been described by others in detail [110–112]. For measuring the contact angle on a flat mineral surface the most suitable
technique is the Axisymmetric Drop Shape Analysis Profile (ADSA-P) method. For measuring the contact angle of real mineral surfaces, the use of capillary penetration methods have been found to be suitable [113], where the contact angle can then be calculated using the Washburn equation [114]. Atomic Force Microscopy (AFM) has proved to be a useful technique for measuring the interactions between bubbles and individual colloidal particles [20,38,115,116]. AFM has been used to measure both the advancing and receding contact angles for individual colloidal particles. Determination of the contact angle using AFM assumes that both the particle and bubble have a spherical profile and by using the experimentally obtained force distance curve, shown in Figure 18, to determine the penetration depth, $D$, of the particle into a bubble, the contact angle can be estimated, using the relation $\cos \theta = \left( R_p - D \right) / R_p$. However, AFM has proved more useful for measuring interactions during the approach cycle rather than the detachment cycle, due to the differences between the cycles [38,39].

![AFM principle of the measurement of contact angles on colloidal particles](image)

**Figure 18.** AFM principle of the measurement of contact angles on colloidal particles [115]

5 Hydrodynamic forces

5.1 Water velocity around air bubbles

The liquid flow around a rising gas bubble is governed by both the continuity and Navier-Stokes equations [117]. Exact solution can be derived analytically for situations when the bubble Reynolds number is infinitely large (potential flow), or equal to zero (Stokes flow). For bubble Reynolds numbers between these limits the continuity and Navier-Stokes equations need to be numerically solved [35].
Figure 19. Notations for liquid flow around a rising bubble, with a velocity, $U$. Spherical coordinates $(r, \varphi, \phi)$ are used to describe the axisymmetric velocity components around the bubble [42].

Considering a rising bubble, we can assume rotational symmetry around the bubble, based on a spherical coordinate system as defined in Figure 19. This simplification allows the continuity and Navier-Stokes equations to be simplified using the convenient axisymmetric spherical coordinate system [35].

The continuity equation can be written as:

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 w_r \right) + \frac{1}{r \sin \varphi} \frac{\partial}{\partial \varphi} \left( w_\varphi \sin \varphi \right) = 0$$

(27)

The corresponding Navier-Stokes equations can also be simplified, resulting in:

$$w_r \frac{\partial w_r}{\partial r} + w_\varphi \frac{\partial w_\varphi}{\partial \varphi} - \frac{w_\varphi w_r}{r} = -\frac{1}{2} \frac{\partial p}{\partial r} + \frac{2}{Re} \frac{1}{r^2} \left( r^2 \nabla^2 w_r - 2w_r - 2w_\varphi \cot \varphi - 2 \frac{\partial w_\varphi}{\partial \varphi} \right)$$

$$w_r \frac{\partial w_r}{\partial r} + \frac{w_\varphi}{r} \frac{\partial w_\varphi}{\partial \varphi} + \frac{w_r}{r} = -\frac{1}{2} \frac{\partial p}{2r} + \frac{1}{Re} \frac{1}{r^2} \left( r^2 \nabla^2 w_\varphi - w_\varphi \frac{w_r}{\sin^2 \varphi} + 2 \frac{\partial w_r}{\partial \varphi} \right)$$

(28)

where the Laplace operator is defined as:

$$\nabla^2 = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial}{\partial r} \right) + \frac{1}{r^2 \sin \varphi} \frac{\partial}{\partial \varphi} \left( \sin \varphi \frac{\partial}{\partial \varphi} \right)$$

(29)

As the fluid flow is considered to be axisymmetric in the Navier-Stokes system of equations a new variable $\psi$ is introduced, called the Stokes Stream Function [118]. Here the velocity components are defined by:
\[ w_r = -\frac{1}{r^2 \sin \phi} \frac{\partial \psi}{\partial \phi}; \quad w_\phi = -\frac{1}{r \sin \phi} \frac{\partial \psi}{\partial r} \]  

Eq. (30) is inserted into Eq. (28), eliminating the pressure term to obtain a single equation, that is satisfied by \( \psi \), [35] resulting in following the fourth order differential equation:

\[
\frac{\partial}{\partial \tau} E^2 \psi + J \left[ \psi, \frac{E^2 \psi}{(r \sin \phi)^2} \right] = \frac{2}{Re} \frac{E^2 (E^2 \psi)}{\sin \phi} \]

(31)

where the Jacobian and differential operators are defined by:

\[
J(a, b) = \frac{\partial a}{\partial r} \frac{\partial b}{\partial \phi} - \frac{\partial b}{\partial r} \frac{\partial a}{\partial \phi} \]

(32)

\[
E^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin \phi}{r^2} \frac{\partial}{\partial \phi} \left( \frac{1}{\sin \phi} \frac{\partial}{\partial \phi} \right) \]

(33)

To assist in the numerical solving of Eq. (31), a vorticity term is included in the differential operator, given by \( E^2 \psi = \Omega \sin \phi \). The vorticity, \( \Omega \) is the name given to the local rate of rotation in a fluid. Using the spherical coordinate system and considering the flow is axisymmetric, the vorticity can be re-written as:

\[
\Omega = \frac{1}{r} \left( \frac{\partial w_\phi}{\partial r} - \frac{\partial w_r}{\partial \phi} \right) \]

(34)

Regardless of whether the bubble surface is considered mobile or immobile the boundary conditions outlined below apply. On the bubble surface the radial velocity \( w_r \) is equal to zero, as water is unable to cross the interface, and \( r = 1 \), yielding,

\[
\psi = 0 \]

(35)

Considering that the fluid flow is unperturbed by the bubble far from the surface \( |\vec{w}| = 1 \) resulting in \( w_r = -\cos \phi \) and \( w_\phi = -\sin \phi \). Inserting these conditions into Eq. (30) and Eq. (35) this results in,

\[
\psi = \frac{r^2 \sin^2 \phi}{2} \]

(36)

\[
\Omega = 0 \]

(37)
The surface mobility of the bubble interface, has been shown to be an important consideration in selecting the boundary conditions to be applied to the surface. In the case of a bubble with an immobile surface, both the normal and the tangential components of the water velocity on the bubble surface asymptotically approach zero [119]. In the case of a bubble with a mobile surface, the surface velocity is not equal to zero [35]. For flotation applications, bubbles with a mobile surface or non-zero tangential velocity component should be considered, as any surface contamination is likely to be swept to the rear of the bubble as it rises through the liquid [120,121].

The water velocity around a mobile bubble surface can be determined numerically by use of a Taylor series expansion [42] resulting for the direction perpendicular to the bubble surface in:

$$\vec{W}_z = -A(z - z^3)\vec{t}_z$$

(38)

where, the water motion in the normal direction to the bubble surface is described by $\vec{w}_z$. Whereas for the case of an immobile bubble surface, the lowest-order prediction available is the second-order model for the normal velocity component of the liquid flow:

$$\vec{W}_z = -A(z^3)\vec{t}_z$$

(39)

In Eqs. (38) and (39), $A$ is a known parameter with a different value for each equation and is a function of the Reynolds number for the front hemisphere of the bubble surface [42]. Note, that first-order predictions have been shown [122] to usually be poor for the modelling of the particle-bubble attachment interactions.

### 5.2 Modified Stokes drag force equation

Stokes’ law governing the motion of a sphere moving slowly through a viscous liquid has a wide range of applications dealing with many different types of sedimentation and fluidisation processes. The well-known relationship, described by Eq. (40), applies only to spherical particles in a fluid medium, which extends to infinity in all directions, classed as an unbounded fluid medium. In many real situations, the presence of other particles, bubbles, rigid walls and free surfaces externally bound the fluid. The presence of these boundaries at a finite distance from the particle under consideration, requires corrections to Stokes’ drag force equation.

$$\vec{F} = -6\pi \mu R_p (\vec{V} - \vec{W})$$

(40)

where $\mu$ is the liquid viscosity, $\vec{V}$ and $\vec{W}$ are the vectors of the particle and fluid velocities, respectively. The Stokes’ drag force will change when the particle approaches another type of surface with an intervening liquid film, where the hydrodynamic resistance increases rapidly [123–126]. This
deviation of the hydrodynamic resistance due to the liquid film and can be accounted for by using hydrodynamic resistance corrections [127]. The Stokes drag force Eq. (40) can be modified to give; in the (radial) direction of the bubble-particle centreline;

\[ F_r = -6\pi \mu R_p V_r f_1 + 6\pi \mu R_p W_r f_2 \tag{41} \]

And in the (tangential) direction perpendicular to the centreline;

\[ F_t = -6\pi \mu R_p V_t f_3 + 6\pi \mu R_p W_t f_4 \tag{42} \]

where the subscript ‘r’ and ‘t’ describe the radial and tangential components of the drag force and the particle and fluid velocities, respectively.

Research into the interaction between a solid particle approaching a solid surface has been well established with all four hydrodynamic resistance functions available as a function of the separation distance between the particle and the solid surface [127–129], following the method set forth by Stimson and Jeffery [123], and others [124,130]. For the interaction between a solid particle approaching an air bubble, the hydrodynamic resistance functions are available as a function of the separation distance: \( f_1, f_2 \)[131], \( f_3 \)[132] and \( f_4 \)[133].

Research into the hydrodynamic resistance functions for a spherical particle floating at a gas–liquid interface, has primarily focused on studying the motion of a particle translating an interface in a stationary flow field. Thus, the drag force correction arising from the particle motion, \( f_3 \), has been well studied and a variety of experimental and theoretical have been undertaken [134–141]. Some of the key results from various studies are shown in Figure 20.

![Comparison of experimental and theoretical Stokes drag force correction coefficients for particle motion across a stagnate fluid-fluid interface.](image)

**Figure 20.** Comparison of experimental and theoretical Stokes drag force correction coefficients for particle motion across a stagnate fluid-fluid interface.
The stability of stationary particle floating at an interface with a defined contact angle in an extensional flow field has been numerically studied by Stoos and Leal [142]. The problem addressed in their work considered the case where an attached particle is swept to the rear stagnation point of a bubble and is exposed to an axisymmetric straining flow tending to detach the particle from the interface. Stoos and Leal [142] adopted a numerical treatment to the problem using a boundary-integral technique to consider the deformation of the interface in the presence of a steady uniaxial extensional flow, presenting the results as a set of dimensionless critical capillary numbers for system stability. The data was obtained through trial and error calculations.

6 Concluding remarks

This review has covered the current theories which describe the stability of floating particles at an gas-liquid interface. Some of the important concepts in relation to the contact angle and its importance for successful flotation have been discussed. The key bubble-particle interactions that occur during the flotation process have also been covered. Particular importance is placed on the current understanding of the detachment process in relation to coarse particle flotation, as it is the rate limiting mechanism. The modified Stokes drag force equation has also been introduced, in particular the implications for the perturbation to the flow around rising bubbles caused by a particle attached to the bubble surface.

Overall, the literature does not describe a clear mechanism for bubble-particle detachment occurring in the pulp phase of a flotation cell. It indicates that it could arise as a result of either bubble deformation/oscillation, centrifugal force or kinetic energy but does not arrive at a definitive conclusion as to which mechanism or combination thereof is responsible for the detachment of coarse particles. Technological advancements in coarse particle flotation utilise fluidisation principles to achieve flotation of a coarser sized particles, but the theory of detachment by centrifugal force is not valid for these emerging technologies. The detachment of particles from an air-water interface has historically been based on a thorough understanding of the capillary theory of flotation, while the contact angle hysteresis as a particle undergoes dynamic detachment from an air bubble has received little attention. Solutions for the Stokes drag force correction for a particle translating across an stagnate fluid-fluid interface, $f_3$, are available in the literature, the remaining solutions for the corrections $f_1$, $f_2$ and $f_4$ or experimental results, are not readily available in the literature.
References


Chapter 3

Effect of shear flow on a spherical particle on detachment from a bubble surface

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Abstract

Detachment of particles from the bubble surface plays an important role in the efficiency of flotation separation of coarse hydrophobic particles from hydrophilic ones in water using air bubbles. The literature indicates that coarse particle flotation depends strongly on quiescent hydrodynamic conditions thereby minimising the probability of bubble-particle detachment. This paper is concerned with the detachment of coarse particles from a stationary air bubble surface in a well-defined shear flow field. The aim is to examine the magnitude of the hydrodynamic forces of the water shear flow on the detachment of a particle (200-500 µm) attached to a captive air bubble. A new experimental technique was developed using a small enclosed channel with fully developed laminar flow at intermediate Reynolds number. Pure water flows past a partial bubble segment protruding from the channel wall. Hydrophobic ballotini were attached to the air bubble and the water velocity increased capturing the particle movements using high speed video microscopy. Observations of the particle movements attached to the stationary bubble are analysed and interpreted and the implications for coarse mineral flotation are discussed.

1 Introduction

Froth flotation has been widely used for over a century to separate valuable mineral particles from gangue particles. The first stage of a beneficiation process using flotation is to grind the ore to liberate (release) the valuable mineral particles from the gangue or waste minerals. Chemical reagents called collectors are added to the ground slurry rendering the exposed surfaces of the valuable minerals hydrophobic. Frothing agents are added to the slurry to facilitate formation of stable bubbles. The slurry is transferred to a suitable cell or column and the particles are suspended and aerated, where upon the hydrophobic mineral particles preferentially attach to the air bubbles. The loaded air bubbles rise to the top of the cell where they are collected as a concentrate product. Particle size has an important role in flotation. Commercially the largest particles that can be floated are around 10 times smaller than that of the particles that can be theoretically floated. This was demonstrated by Gaudin et al. [1] under quiescent conditions for sulphide minerals (2-3 mm) and for bituminous coal (10.7 mm). The recovery of minerals by flotation is typically most successful in the 10-200 µm size range. In order to optimise flotation performance, it is important to understand the forces acting on particles attached to an air bubble as well as the mechanism by which the particle are detached from the surface of the air bubble.

Coarse particles are well known to have a poor recovery in conventional flotation cells. Part of this is due to a decrease in the proportion of liberation of the floatable mineral as the particle size increases. However, in the literature there are numerous reports of coarse, fully liberated particles
being present in flotation plant tailings, indicating that liberation is only part of the puzzle. Various studies and theories have been undertaken to identify the mechanism of detachment in relation to coarse particle flotation [2–5]. The literature generally suggests that the detachment of bubble-particle aggregates arises due to the high-turbulence present in a mechanical flotation cell and during the transfer of heavy particles from the pulp to the froth phase.

Considering the transfer of particles from the pulp into the froth, a mechanism was proposed by Falutsu and Dobby [6] that the sudden deceleration of the bubble-particle aggregates as they collide with the froth phase is the cause of detachment. Recently Ata [7] showed that the bubble surfaces oscillate as the loaded bubbles collect and coalesce in the froth phase. The oscillation of the bubble surface is an important detachment mechanism in detaching the less hydrophobic particles.

In the pulp phase of a mechanical flotation cell Woodburn et al. [8] proposed that bubble deformation is the mechanism of particle detachment. They suggested that this is due to the tension stress applied on the bubble by sudden acceleration of the aggregate. If this stress exceeds a certain value the bubble surface fails and the particles detaches. Holtham and Cheng [2,3] measured bubble-particle detachment forces using a vibration technique, finding that the amplitude of oscillations imposed on the bubble is a dominant factor in the detachment process for the specialised system. This technique was further developed by Xu et al. [9] who examined the detachment of coarse particles from oscillating bubbles as a function of particle hydrophobicity and shape, as well as medium viscosity. This technique of measuring the detachment force was also used in the analysis for various recent studies [10,11].

In his series of publications Schulze [5,12,13] developed a theoretical prediction for coarse particle detachment and postulated that the force of detachment was due to the centrifugal force that is developed when a bubble-particle aggregate is located in a turbulent eddy. It was assumed that the eddy size was equal to the bubble diameter and he used Kolmogorov’s isotropic turbulence theory to determine the rotational speed of the eddy, allowing the calculation of the centrifugal force exerted on the particle. Goel & Jameson [14] carried out an experimental study of bubble-particle detachment in a specially designed flotation cell to capture the effect of bubble-particle detachment in an eddy and test the centrifugal effect on particle detachment. They found that the detachment of particles follows a distribution of predicted values rather than a critical number.

Another mechanism of detachment in the pulp phase was first proposed by Yoon and Mao [15] describing that bubble-particle detachment will occur if the particle exceeds a critical level of kinetic energy, which is calculated from the particle motion in relationship to the bubble. This theory has been expanded by theoretical modelling work by Gontijo et al. [16] and Wang et al. [17] with the slight modification that the amount of kinetic energy is calculated from the velocity of the bubble-particle aggregate moving through the pulp.
Overall the literature does not describe a clear mechanism for bubble-particle detachment occurring in the pulp phase of a flotation cell. It indicates that it could arise as a result of either bubble deformation/oscillation, centrifugal force or kinetic energy and does not arrive at a definitive conclusion as to which mechanism or combination thereof is responsible for the detachment of coarse particles.

In this paper a new and novel experimental technique has been developed to determine the shear force required to detach a particle from a captive air bubble. This has been achieved using a small enclosed channel with fully developed laminar flow at intermediate Reynolds number. The water flows past a bubble segment protruding from the channel wall. Spherical hydrophobic ballotini were attached to the air interface and the water velocity increased capturing the particle movements and detachment using high speed video microscopy. The observations were analysed and interpreted in terms of coarse particle detachment theory.

2 Experimental

A specially constructed flow channel that allows observation of particle detachment from a captive air bubble was constructed from 3 mm think cast poly methyl methacrylate (Perspex). The device was formed from four pieces of the acrylic, with the internal channel dimensions machined to be 5 mm square and 80 mm long. For creating a bubble in the channel a 1 mm hole was carefully drilled through one of the pieces, in a location to allow for side on observation of the captive air bubble. To form the device into a complete channel the pieces were joined together using a specialty acrylic glue (ACROFIX IR 0192), details shown in Figure 1.

The particles used to observe detachment were spherical glass ballotini. The ballotini were left untreated in order to obtain a range of contact angles. The particulate size of the ballotini was in the range 200-500 µm, this is the size range typically considered to be coarse in conventional flotation cells. The water used in the experiments was freshly purified using a reverse osmosis RIO’s unit and an Ultrapure Academic Milli-Q system (Millipore, USA).
Using the arrangement illustrated in the water is pumped from a reservoir, using a brushless 12 VDC centrifugal pump, up to a constant head overflow tank. Water flows from the head tank under gravity through the acrylic device, the flow rate through the system is controlled using a downstream ball valve. The size of the air bubble was manipulated using a Micro 4 syringe pump controller (World Precision Instruments, USA) via the 1 mm hole in the channel wall. A high speed camera microscope camera (Photron Fastcam SA1.1, USA) was used to record images of the bubble-particle aggregate. A fibre light with diffuser was used to illuminate the droplet from behind to provide a high contrast image.

3 Results and discussion

3.1 Contact angle measurement

As the glass ballotini surfaces were left untreated, the contact angle was calculated for each particle once it was attached to the captive bubble, Figure 2 (a) is an example image that was used to digitise data of the particle and bubble circumferences. The digitised data was then used to determine the constants $a_1$, $a_2$ & $a_3$ for the equation of best fit $x_2+y_2+a_1y+a_2y+a_3=0$ for both the bubble and particle. The constants from the equations for the bubble and particle were used to calculate the angle between the respective tangents at the point of intersection of the two circles - this angle is assumed to be the contact angle based on no deformation of the interface, as the particle is much smaller than the bubble (see Figure 2 (b)).
Figure 2. Image used to calculate the particle contact angle (a) a particle with a contact angle of 39°±1°, particle diameter 307 µm. (b) Sketch of the image (not to scale) showing the tangents at the point of intersection and the angle created.

3.2 Water flowrate measurement and calibration

Determination of the volumetric flowrate past the captive bubble was determined by a series of timed volume collection tests. The calculated volumetric flowrate was correlated to the angular position of the ball valve the results are plotted in Figure 3. The data was fitted to a polynomial equation with an R² value of 0.998 to help overcome some of the positioning issues with the ball valve. The equation was used to determine the volumetric flowrate from the valve position.

Figure 3. Valve calibration chart

3.3 Influence of bubble shape

Initial experiments were conducted with a 1.3 mm diameter bubble protruding into the flow channel (see Figure 4). From these tests it was found that at higher flow rates the particle is swept to
the rear of the bubble becoming trapped in the low pressure region formed. The formation of a low pressure wake was identified from videos of the particle shown in the last two images of the sequence in Figure 4. The videos revealed that the particle slides around the rear section of the bubble surface. In the last image, when the water flowrate has been increased to 31 L/h the bubble has begun to detach from the tube with the particle still remaining intact.

**Figure 4.** Full size bubble in increasing flow regime, fluid flow direction is from the left to the right hand side (particle diameter 360 µm, bubble diameter 1.3 mm, contact angle 32°)
3.4 Particle detachment sequence

To avoid the formation of a low pressure region at the rear of the bubble, the air interface was manipulated to be level with the channel wall. This method positions the particle to be exposed directly to the force of the water applied, without creation of a low pressure region. Using this method particle detachment from the air water interface was able to be accurately observed. Figure 5 show the sequence of images for the slow increase of the water flowrate leading to eventual particle detachment.

![Image](image_url)

**Figure 5.** Sequence of particle detachment in increasing flow regime, particle diameter 318 µm and contact angle 34°. Particle detaches at a water flowrate of 26.6 L/h, this equates to a water velocity of 5.5 cm/s and $\text{Re}_p$ 17.

In Figure 5 the last five images show the frames captured in the instance leading to detachment. Analysis of the video indicates no oscillating motion of either the bubble surface or particle, rather the particle appears to be suddenly tugged off the air surface. Below each of the image frames captured is listed the average channel volumetric flow rate and water velocity acting at the particle centre at the time of capture.
Reducing the profile of the bubble surface had the added benefit of simplifying the flow profile in the channel. The fluid velocity acting at the centre of the particle $V_x$ can be calculated using a rough linear approximation assuming that the particle is much smaller than the boundary layer (Eq. (1)).

$$V_x = 6\left(\frac{Q}{A}\right)\left(\frac{x}{l}\right)\left(1 - \frac{x}{l}\right)$$

where $Q$ is the volumetric flowrate, $x$ is the vertical distance from the channel surface, $A$ is the cross sectional area of the channel, and $l$ is the channel width.

We can now plot the point of detachment as a function water velocity acting at the particle centre and contact angle measured (Figure 6). The data, while not providing a clear correlation between the two parameters, indicates that there is a trend that with increasing contact angle a higher water velocity is required for detachment to occur.

![Figure 6. Contact angle as a function of water velocity at the point of detachment.](image)

3.5 Force of adhesion

The strength of the particle attachment can be quantified using a force balance [18] i.e., a balance of the adhesive forces to detaching forces. The particle will detach from the air-water interface only if the detaching force exceeds the tenacity of attachment.

The simplification of the force balance has been condensed into Eq. (2) [19]. The equation incorporates the adhesive forces of capillary, hydrostatic pressure and the buoyancy of the particle volume immersed in the liquid, along with the detaching force of the particle weight.

$$T = \pi R_p \sigma (1 - \cos \theta) \left[1 + 0.016 \frac{R_p}{L}\right]$$

(2)
where $T$ is the force of attachment, $L = \sqrt{\sigma / \delta g}$ is the capillary length, $\theta$ is the three phase contact angle, $R_p$ is the particle radius, $\delta$ is the water density, $g$ is the acceleration due to gravity, and $\sigma$ is the liquid surface tension.

The calculated attachment tenacity has been plotted as a function of the water velocity, as shown in Figure 7.

![Figure 7. Attachment force as a function of water velocity at the point of detachment](image)

The behaviour of a fluid flow can be characterised using the well-known dimensionless Reynolds number. The particle Reynolds number can be defined in a similar way, as in Eq. (3), which describes the behaviour of fluid flow around the particle.

$$Re_p = \frac{2 \delta R_p w}{\mu}$$  \hspace{1cm} (3)

where $w$ is the water velocity at the particle centre calculated using Eq. (1) and $\mu$, the liquid viscosity. For the detachment experiments conducted the calculated Reynolds numbers for water flow around the particle are in the range between 15 and 30. This means that the flow, disturbed by the presence of the particle, can be described by creeping flow and therefore the flow can be described in terms of a Stokes stream function. This then allows for analytical solutions of the Biharmonic differential equation of the disturbed flow to be developed. The solutions can be used to model the hydrodynamic force for the fluid pressure exerted on the particle attached to an air water interface, leading to an exact analytical expression for a Stokes drag force correction factor.

4 Conclusions and recommendations

In this work, an experimental setup was designed and built for investigating the detachment of submillimetre ballotini particles from the bubble surface in a shear flow field of water. The particle-
bubble detachment was captured using high-speed video microscopy and quantified in terms of the particle hydrophobicity (contact angle) and the local water velocity. It was found that a quantifiable hydrodynamic force was able to detach the particles from the bubble surface. The hydrodynamic detachment of particles cannot be explained on the basis of oscillation of the bubble surface or particle. The stability of spherical particles attached to the gas-liquid interface and its subsequent detachment can be analysed based on a force balance. The results indicate that there is a trend for the detachment of the particles exhibiting a higher level of hydrophobicity, as indicated by the contact angle, requiring an increased velocity for detachment to occur. It is anticipated that the results obtained can be used for interpreting the flotation results of coarse particle and for modelling bubble-particle detachment efficiency.
References


Chapter 4

An analysis of bubble deformation by a sphere relevant to the measurements of bubble-particle contact interaction and detachment forces

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Abstract

Atomic force microscopy makes it possible to measure the interacting forces between individual colloidal particles and air bubbles, which can provide a measure of the particle hydrophobicity. To indicate the level of hydrophobicity of the particle the contact angle can be calculated, assuming that no interfacial deformation occurs with the bubble retaining a spherical profile. Our experimental results obtained using a modified sphere tensiometry apparatus to detach sub-millimetre spherical particles show that deformation of the bubble interface does occur during particle detachment. We also develop a theoretical model to describe the equilibrium shape of the bubble meniscus at any given particle position, based on the minimization of the free energy of the system. The developed model allows us to analyse high-speed video captured during detachment. In the system model deformation of the bubble profile is accounted for by the incorporation of a Lagrange multiplier into both the Young-Laplace equation and the force balance. The solution of the bubble profile matched to the high-speed video allows us to accurately calculate the contact angle and determine the total force balance as a function of the contact point of the bubble on the particle surface.

1 Introduction

Interactions between liquid-gas interfaces and solid particles are of particular interest in a number of industrial separation processes. One example is froth flotation, which is a unit process widely used in the recovery of valuable base, precious and industrial minerals. During flotation, separation is achieved by the selective attachment of hydrophobic particles to air bubbles. Hydrophilic particles do not attach to the air bubbles and remain in the slurry to be discarded.

An important measure of a solid surface hydrophobicity is the three-phase contact angle. To measure the contact angle on flat surfaces, the sessile drop technique can be simply used. However, for particulate solids, measurement of the contact angle is far more complex. A common technique performed utilizes the capillary rise technique through a packed bed of particles determining the contact angle by solving the Washburn equation [1].

The analysis of forces exerted on particles attached to a bubble surface has been and continues to be a central focus of research into froth flotation [2–4]. The principle of particle stability at the air-water interface has been used to determine the surface tension [5,6] for various liquids, it has also
been confirmed experimentally by using centrifugal forces [2,7] to detach particles. Particle detachment has also been studied using an acoustic technique [8,9] to oscillate bubble-particle aggregates and thus determine the detachment force required to detach the particle from a stationary bubble.

Interactions and film drainage between bubble and particle surfaces has been investigated extensively by others [10] and recently using an interfacial thin film drainage apparatus (ITFDA)[11]. Work performed by Wang et al. [12] on the approach of bubble to a hydrophobic sphere found that for low bubble deformation, the calculated and measured forces were shown to agree well with each other but not in the case when large bubble deformation occurred.

Atomic Force Microscopy (AFM) has proved to be a useful laboratory technique for measuring the interactions between bubbles and individual colloidal particles [13–16]. AFM has been used to measure both the advancing and receding contact angles for individual colloidal particles. However, AFM has proved more useful for measuring interactions during the approach cycle than the detachment cycle [14,17]. Determination of the contact angle using AFM is completed by identifying the zero force position in the contact region from the force distance curve and assuming that both the particle and bubble have spherical profiles.

![Figure 1](image.png)

**Figure 1.** Illustration of the vertically resolved component of the capillary force on the particle. $\theta$ is the particle contact angle, $\alpha$ is the central angle, $\beta$ is the angular inclination of the air-water interface at the three-phase contact point, and $\sigma$ is the air-water interfacial surface tension [17].

Using the force distance curve the contact angle, $\theta$, can be simply calculated using the equation $\cos \theta = \left( \frac{R_p - D}{R_p} \right)$, where $R_p$ is the particle radius, and $D$ is the relative depth of penetration of the particle into the bubble, identified from the position of the zero force [14]. This method relies on the key geometrical assumption that the bubble meniscus profile remains spherical during the approach and detachment cycles. The vertically resolved capillary force [4] on the particle, $F_\sigma$, as shown in Figure 1, is given by $F_\sigma = 2\pi\sigma R_p \sin \alpha \sin (\theta - \alpha)$. At the zero force position the
term \( \sin(\theta - \alpha) \) should equal zero, allowing the contact angle to be determined by equating the contact angle to the central polar angle of the particle. It is well known [16,17] that there is some hysteresis in the point of zero force between the approach and retraction parts of the force distance curve in the contact region.

The amount of bubble deformation occurring during particle detachment using AFM is difficult, if not impossible, to accurately measure, due to the size scale used in AFM. The consequence of this is that the detachment cycle cannot be accurately described using this methodology, as it is not possible to unambiguously start with the total force balance and calculate the shape of the bubble profile and surface forces[14,18].

Recent work analysing the free energy of particles attached to liquid droplets by Guzowski et al. [19] and Ettelaie & Lishchuk [20] calculated the deformation profile of the droplet from a force that is applied to particles attached at the interface. In both these studies, they considered the three phase contact line to be freely moving over the particle surface taking the contact angle to be an unchanging system constant. Alternatively, recent studies [16,21,22] have considered the case where the contact line has been pinned at a sharp edge on a particle surface. Using particles with circular cuts in a study applying AFM conducted by Ally et al. [16], their work clearly showed that the contact angle at the edge could take a range of values within the range defined by the Gibbs inequality condition.

The work in this paper seeks to understand why there is hysteresis of the contact angle between the approach and retraction of a particle using AFM type techniques and further the extent of bubble deformation. In the experimental work completed using a modified sphere tensiometry apparatus, the force exerted on a particle during detachment from an air bubble has been measured. The approach and detachment cycles were observed using high-speed microscopy to record changes in the bubble profile as the particle underwent detachment.

As has been shown in the literature [23,24] undertaking a force balance for complex capillary systems can lead to errors. Another more rigorous approach for describing the stability of bubble-particle aggregates can be performed by minimization of the free energy of the capillary system. Using the energy minimization technique, we present here a new theoretical model to describe the profile of a deformed sessile interface by two objects and calculate the forces exerted on the particle in question, based on the system geometry. Importantly, the problem to be solved is different here from previous methodologies [16,19,20,22,25] in that in this work, we consider the case for which the contact line is not pinned at a sharp edge and where the contact angle is not constrained as a constant.
## Theoretical Modelling

In this section, we develop a theoretical model based on analysis of the free energy of the bubble-particle system to describe the deformed bubble profile, given the unconventional deformation of the bubble which is attached to the flat surface and at the same time being stretched or pressed by the particle under applied force (Figure 2). The equilibrium shape of the bubble at any given particle position can be obtained by minimising the free energy of the system. The novel aspect of this model that the contact line is considered not to be pinned and that the three-phase contact angle is not used as a system parameter. The energy minimisation is carried out as a function of the Three-Phase Contact (TPC) position which is determined by its height, \( h_{tpc} \) and radial coordinate, \( r_{tpc} \). It is noted that the TPC position is also a function the height of the particle centre, \( h_c \). However, the heights of the TPC position and the particle centre are coupled via the central angle, \( \alpha \), of the TPC position via \( h_c = h_{tpc} + R_p \cos \alpha \). Therefore, the energy minimisation can be written as a function of \( h_{tpc} \) and \( \alpha \) (and the radius, \( r_0 \), of the bubble contact with the flat solid surface). Using these parameters, we are able to capture any changes to the value of the contact angle throughout the interaction of the particle with the bubble.

![Figure 2](image_url)

**Figure 2.** Schematic of a bubble attached to both a spherical particle and a solid planner surface. \( r_c \) is the contact radius on the particle surface, \( r_o \) is the bubble contact radius, \( h_{tpc} \) is the height of the meniscus connected to the three phase contact line, \( R_p \) is the particle radius, and \( h \) and \( r \) describe the coordinate system.

To minimise the free energy, \( E \), of the capillary system under consideration (shown in Figure 2) we need only to minimise the gravitational potential energies and the interfacial energies of the system to obtain the shape of the meniscus. The bubble volume, \( V_b \), is considered to be constant for
this system; this constraint is included in the functional for minimisation by the use of a Lagrange multiplier, $\lambda$. The resulting functional to be minimised can be written as:

$$E = E_{\text{volume}} + E_{\text{surface}} - \lambda V_b$$  \hspace{1cm} (1)

The details for completing the minimisation of the free energy for this system can be found in Appendix A. The results of the analysis yield three key equations: the Young-Laplace equation, the balance of forces on the particle, and the Young equations. The Young-Laplace equation derived in this work is different from traditional representations [26] as it includes the Lagrange multiplier to meet the bubble volume constraint. The Young-Laplace equation is described as follows:

$$\sigma \left\{ \frac{d^2 h / dr^2}{\left[ 1 + (dh / dr)^2 \right]^{1/2}} + \frac{dh / dr}{r [1 + (dh / dr)^2]^{1/2}} \right\} = \Delta \rho g h + \lambda$$  \hspace{1cm} (2)

where $\Delta \rho = \rho_l - \rho_a$ is the positive difference between the liquid and air densities, $g$ is the acceleration due to gravity, and $\sigma$ is the liquid surface tension.

The completion of the minimisation of energy also provides a useful balance of forces on the spherical particle. For simplicity, only the particle weight is considered in the analysis in the SI. The externally applied force on the particle in our experiments can be accounted for in the analysis by adding it to the particle weight. If the external force, $F_{ex}$, is applied onto the particle in the direction of gravity, the force balance given the SI can be modified to yield:

$$mg \left( 1 - \frac{\delta}{\rho} \right) + F_{ex} = -2\pi \sigma r_{pc} \sin \beta + \pi r_{pc}^2 \left[ h_{pc} g \Delta \rho + \lambda \right] - \pi g \Delta \rho R^3 \frac{2 - 3 \cos \alpha + \cos^3 \alpha}{3}$$  \hspace{1cm} (3)

where $m$ is the mass of particle, $\rho$ is the particle density, $R_p$ is the particle radius and $\beta$ is the angle of meniscus inclination to the horizontal line at the TPC location. The last term on the right hand side of Eq. (3) is the weight of liquid in air, which has the volume of the spherical cap of particle immersed in the bubble. The first term on right hand side of Eq. (3) is the vertical component of the capillary force on the wetting perimeter, and second term is the bubble pressure force on the gas-solid contact area.

As at the beginning of each of the experiments the force sensor with the attached particle in water was off-set to zero and the overall change in the height of the immersed section of the wire was negligible, Eq. (3) for the measured force can be reduced to:

$$F_{ex} = -2\pi \sigma r_{pc} \sin \beta + \pi r_{pc}^2 \left[ h_{pc} g \Delta \rho + \lambda \right] - \pi g \Delta \rho R^3 \frac{2 - 3 \cos \alpha + \cos^3 \alpha}{3}$$  \hspace{1cm} (4)
where a positive value for $F_{ex}$ indicates the external force is acting in the direction of gravity (pushing the particle toward the bubble), and when negative the external force is acting against the direction of gravity (pulling the particle away from the bubble).

Equation (2) can be numerical solved to obtain the bubble interface profile which can be compared with the experimental profile of the bubble shape to quantify key parameters, including $\lambda$, $\beta$, $\alpha$ and $\theta$. The differential equation can be converted to a solvable parametric equations by using the arc length, $s$, of the meniscus [27], yielding:

$$\frac{dx}{ds} = \cos \phi$$

$$\frac{dy}{ds} = \sin \phi$$

$$\frac{d\phi}{ds} = y + \lambda^* - \frac{\sin \phi}{x}$$

(5)

where $\phi$ is the angular inclination of local bubble meniscus to the $r$-axis, i.e. $\frac{dh}{dr} = \tan \phi$, and $x = r/L$ and $y = h/L$ are the dimensionless variables, $s$ is the dimensionless arc length of the meniscus, $L = \sqrt{\sigma / (g \Delta \rho)}$ is the capillary length, and $\lambda^* = \lambda / \sqrt{\sigma g \Delta \rho}$. The set of the parametric equations can be integrated using the fourth-order Runge-Kutta scheme and the boundary conditions at the TPC position, i.e., $x = x_{tpc}$ and $y = y_{tpc}$ at $\phi = \pi - \beta$.

The numerical solution to the Young-Laplace equation can also be used to provide additional information for the bubble volume, $V_b$, and the surface area, $A_b$, of the bubble, which are defined as

$$V_b = \pi \int_0^{s_{max}} r^2 \sin \phi \, ds - \pi R_p^3 \frac{2 - 3 \cos \alpha + \cos^3 \alpha}{3}$$

(6)

$$A_b = 2\pi \int_0^{s_{max}} r \, ds$$

(7)

Equations (21) and (22) were integrated simultaneously together with Eq. (20) using the fourth-order Runge-Kutta scheme.

3 Experimental Methods

3.1 Materials

The particles used to observe detachment were sub-millimetre spherical borosilicate glass beads (Sigma-Aldrich, Australia) with a diameter of approximately 500 $\mu$m. The model particles were hydrophobised by esterification in 1-octanol [28–31] to obtain contact angles of around 85°. The
silica particles were cleaned ultrasonically in ethanol and water then dried in a desiccator. The silica particles were esterified using 1-octanol for 4 h and washed ultrasonically with acetone. The particles were glued onto a thin wire (820 µm) using a small amount of epoxy resin (Selleys Araldite Super Strength, Australia). The water used in the experiments was freshly purified using a reverse osmosis RIO’s unit and an Ultrapure Academic Milli-Q system (Millipore, USA). The surface tension of the water was taken as 0.072 N/m and this value was adopted for use in the calculations.

3.2 Experimental

The experimental setup is shown Figure 3, it comprises of a force balance (XS205 Dual Range Analytical Balance with 0.01 mg accuracy, Mettler Toledo, USA), a motorized laboratory jack (L490MZ/M Motorised Lab Jack, Thorlabs, Cambridgeshire, UK), a rectangular liquid cell (clear acrylic) for containing the liquid and a Teflon base plate, a spherical test particle mounted to the bottom of the force sensor, a high-speed microscope camera video (Photron Fastcam SA1.1, USA), and a PC for synchronising the motion of the motorised stage and recording the force measurement data.

**Figure 3.** Experimental setup, not to scale, consisting of a force balance, a motorized lab jack, a liquid cell, suspended test particle, a high-speed camera with background lighting, and a PC for synchronising and data collection.

The rectangular acrylic cell containing a Teflon base plate was placed on the laboratory jack used for vertical translation. Both the force balance and laboratory jack were operated by Lab View software developed in-house and installed on the PC to synchronise the vertical lab jack displacement and the force measured by the balance (The force data was recorded every 100 ms where the high speed camera captures a frame every 4 ms. To resolve the difference in the rates of data collection by the force balance and the high-speed camera, a spline (monotonic) algorithm that has removed potential artefacts and corrected the steps in the force diagrams was implemented). The cell was filled with Milli-Q water. A small bubble was placed on the Teflon plate using an Eppendorf pipet. To ensure that the apex of the bubble was located centrally below the suspended particle, ensuring the
system was rotationally symmetric, the fluid cell was placed on an x-y table attached to the lab jack. A second microscope camera was positioned perpendicular to the high-speed camera. Then, using a fine grid on the video display for both cameras, the x-y table was manipulated locating the bubble centrally beneath the particle. To minimise vibration and disturbances, the whole setup was placed on a vibration isolation table in an air-conditioned laboratory at room temperature of 23°C. The force sensor and the motorized stage were enclosed in a clear acrylic box to isolate the effect of any local airflow on the motion of the test particle and the air-liquid interface.

![Figure 4. An example of the recorded force data measured and the lab jack position as a function of time. Note the force data has been shifted 920 ms to the left to account for signal latency.](image)

The upward and downwards speed of the motorised stage was 0.01 mm/min and the particle was held in a stationary position for five seconds (see Figure 4) before the lab jack was retracted detaching the particle.

### 3.3 Image analysis

Each frame of the video recorded by the high-speed camera beginning from particle attachment until particle detachment was analysed to determine the particle contact angle, central angle, TPC particle contact radius, and meniscus height. Using the built in Canny edge detection method supported by the `edge` function in MATLAB’s Image Analysis Toolbox, a binary image of the edge profiles was generated. The individual outline profiles for the Teflon base, wire, particle and bubble were determined. The image was rotated to align the bubble profile in the direction of gravity; the angle was determined using the `procrustes` function in MATLAB Statistics and Machine Learning Toolbox. The dimensions of the bubble, particle and relative base position were determined by finding the ratio of pixels to length in µm, calculated based on the known diameter of the wire. The origin of the coordinate system to solve Eq. (2) was determined by fitting the particle profile to an equation of a circle. The particle centre was defined as the vertical axis and the position of the Teflon base, set against the horizontal axis origin.
The fitting parameters $\lambda^*$ and $\beta$ used to define the boundary conditions to model the deformation profile, Eq. (20), were determined using the *lsqcurvefit* nonlinear least-squares solver function in MATLAB’s Optimisation Toolbox. The solver was implemented using the trust-region-reflective algorithm. Optimisation of the problem was performed using multiple start points, to ensure a global minimum was reached.

![Fitted bubble profile](image)

**Figure 5.** Fitted bubble profile. The particle size measured from the image is 530 ± 2 µm.

To correlate the timestamps for the high-speed camera video and the displacement data, the calculated position of the bubble base from each video frame was scaled to match the dimensions of lab jack position data. Two displacement versus time curves were created, and the video timestamp was offset by a given factor to overlay the two displacement curves (see Figure 6).
To account for the signal latency between the recorded position of the lab jack and the force data collected, an offset factor was applied to the force data time-stamp. The factor was determined by calculating the variance of the force data, over the stable particle position. The variance was then minimised by adjusting a time offset factor, based on the assumption that if the lab jack is stationary, the force remains constant. To assign a measured force value to each frame of the high-speed video, a cubic spline interpolation algorithm was implemented using the corrected time stamps to correlate the data for the measured force exerted on the particle.

4 Results and Discussion

4.1 Experimental results

4.1.1 Video sequence

The bubble-particle interactions captured by the high-speed camera as shown below in Figure 7. The results shown in this and the previous sections are typical of the other experiments conducted and are of a single run. The sequence of images shows the approach cycle, frames 1 to 3, as the bubble is first lifted into contact, frame 2, with the particle. Frame 4 show the stationary lab jack position before the bubble is lowered. The detachment cycle, frames 5 to 10, show the sequence of bubble-particle detachment as the bubble is pulled away from the particle, detaching in the final two frames.
Figure 7. Key video frames captured during a single experimental run showing the sequence of particle attachment (frames 1-3), the stationary position (frame 4) the particle detachment (frames 5-10). The particle diameter is 530 ± 2 µm and the video was recorded at 250 frames per second for a total of 2,703 frames.

4.1.2 Contact angle variation

The program developed during this study was successfully able to fit the modified Young-Laplace equation described by Eq. (2), to the deformed air bubble profile between two objects. The height of the three-phase contact line is determined from the intercept of the calculated meniscus profile with the circle equation fitted to the particle profile. The contact angle of the particle can then be calculated using the angle of inclination from the solved Young-Laplace equation at the point of intersection. The results of particle contact angle as a function of the meniscus height are plotted in
Figure 8, showing the particle attachment occurring at frame 2 and bubble particle detachment at frame 9.

Figure 8. Experimental results for contact angle vs. meniscus height. Note that the numbers correspond to the frame numbers in Figure 7. (2) Particle attachment occurs, (4) particle held in stationary position and (9) particle detachment occurs.

By considering the particle contact angle as a function of the central polar angle of the actual contact point on the particle, we can gain a better understanding of how the contact angle changes, as the contact line slides over the particle surface and where pinning of the interface occurs.

Figure 9. Experimental results for contact angle vs. angular position. Note that the numbers correspond to the frame numbers in Figure 7. (2) Particle attachment occurs, (4) particle held in stationary position and (9) particle detachment occurs.

As can be seen in Figure 9 there is a steady increase in the contact angle as the bubble is pressed into the particle, frames 1 to 4. During the detachment cycle the bubble returns to a spherical shape, frame 5. The contact line is pinned in between frames 5 and 6 until the contact angle reaches
a certain value where upon the bubble profile begins sliding across the surface again. In frame 6 has
the deformation of the bubble surface is clear. The contact angle increases again in frames 7 & 8 and
detaches in frames 9 & 10.

4.1.3 Volume constraint

To confirm the volume constraint is met, a plot of the calculated bubble volume and the
Lagrange multiplier, \( \lambda \), as a function of time is shown in Figure 10. The plot shows that from particle
attachment through to detachment the bubble volume is constant with a value of around
0.43 \( \pm \) 0.01 \( \mu \)L. The volume was calculated by integrating the calculated bubble profile as a volume
of revolution and subtracting the portion of volume occupied by the particle. The variation of
\( \pm \) 0.01 \( \mu \)L is due to the difficulty of accurately defining the intersection of the bubble with the Teflon
plate from the video frames, as there is a slight angle to viewing the base plate, due to the vertical
translation of the lab jack. The plot of bubble contact radius with the Teflon plate as a function of
time is shown in Figure 11. The data demonstrates that the bubble radius is not fixed but is also
changing as the bubble profile deforms indicating that the contact line is not pinned to the flat plate.

![Figure 10. Calculated bubble volume (blue, o) and the Lagrange multiplier \( \lambda \) (red, \( \Delta \)) as a function of time.](image)
Figure 11. Radius, $r_0$, of the bubble contact with the flat solid surface as a function of time. The contact radius is unchanged during the stop of the lab-jack (cf. Figure 6).

4.1.4 Force data

Figure 12. Experimental force exerted on the particle measured as a function of the position of the lab jack. The force data has been offset to zero at particle attachment.

Typical experimental force data collected as a function of the lab jack position, as shown in Figure 12, was offset to zero at the point of attachment to discount any hydrodynamic forces acting on the particle as the bubble approached the particle. The measured force exerted on the particle was converted to a function of the particle central polar angle, and the typical result of the conversion of the force data is illustrated in Figure 13.
The value of $\lambda$ was calculated for each video frame during the approach and detachment cycle as previously shown in Figure 10. The calculated value of lambda was substituted into Eq. (4) describing the force balance derived through minimisation of free energy. The other parameters including the particle contact radius of the three-phase contact line, the height of the meniscus, the particle central polar angle, and the angle of inclination of the bubble interface were also determined from each frame of the high-speed video captured. The values were used to calculate the force exerted on the particle, the results of which are shown in Figure 14, with particle attachment occurring at frame 2 and detachment at frame 9. Comparing the calculated results using Eq. (8) to the measured forces in Figure 14 show that the theory agrees with the experimental data.

The experimental results outlined here clearly confirm that bubble particle detachment occurs in multiple stages. First, the depressed bubble meniscus returns to a spherical shape. Analysis of the
contact angle and the particle central polar angle in Figure 15 shows that there is only one location where both the angles equal each other, occurring at 38°, and furthermore this angle does not correlate to the point of zero force. The second stage in detachment results in significant deformation of the air bubble as the contact angle increases with partial pinning of the contact line. In the third stage of detachment, the three phase contact line begins to move along the particle surface until the contact angle reaches a critical angle. At this point the capillary system is as described in Figure 2 the particle has a negative penetration depth compared to the original equilibrium shape of the bubble prior to particle attachment.

![Figure 15](image)

**Figure 15.** Comparison of the modelled force (green, o), measured force (red, Δ) and contact angle (blue, ◊) as a function of the particle central polar angle.

The final stage of detachment shows another sudden increase of the contact angle as a distinctive neck forms and detaches from the particle. Frame 10 in Figure 7 shows the instance after detachment, and note that at the base of the particle there is a visible micro-bubble remaining attached to the surface. It is proposed that the bubble presence is due to the necking of the bubble meniscus and detachment occurs as a result of the neck joining. The presence of this bubble was observed in many of the experimental runs and dissipates after a short period of time.

The size scale of used in our experiments where the diameters of our particles are 500 μm and 1.5 mm for the bubbles is highly relevant to coarse particle flotation [32] as we are dealing with the same size scale. The flotation behaviour of coarse particle is predominantly determined by the stability of the bubble-particle aggregates in the pulp phase of a flotation cell and the magnitude of contact angle hysteresis. Our results show the dynamic nature of the contact angle which has important implications for calculating the efficiency of bubble-particle stability in modelling the overall recovery in a froth flotation cell.

AFM measurements typically use smaller particles 20-100 μm and bubble sizes between 0.5-1.0 mm. However, our bubble diameter is still smaller than the capillary length \( L = 2.7 \) mm for the air-water interface. This shows us that the effect of the interface curvature on the bubble-particle...
contact interaction is still significant as in the case of bubbles used in the AFM measurements of the contact and detachment interactions. Also, the bubble to particle diameter ratios of these two systems are still within the same order of magnitude. We believe that this analysis provides an important concept that can be applied in future AFM-based studies that investigate the detachment of particles from bubbles and droplets.

The condition of the pinned contact line has been used [33], but we also found that the condition of the de-pinned contact line has been applied [34]. It is not surprising since it is impossible to see the bubbles in the AFM measurements. Therefore, our paper is of significance regarding this aspect because it provides direct evidence of the depinning condition. Anachkov et al. [25] developed and validated a model using AFM, predicting the location of the TPC at the point of detachment as a function of $\alpha$ for the case of a particle detaching from a flat fluid interface. Application of their model to our work predicts the point of detachment to occur with a value of $\alpha$ between 27-32°. Whereas in our experimental results the point of detachment is observed to occur at central polar angle of 10-15°. This highlights the difference arising to curvature effects of the bubble surface, suggested to have a contributing influence by Preuss & Butt [14] in determining the contact angle based on the point of detachment of a particle from a bubble compared to a flat interface.

5 Conclusions

In this paper the attachment and subsequent detachment of a sub-millimetre spherical particle to an air bubble using, a modified sphere tensiometry apparatus to measure the force, was tracked. The process was recorded using high-speed video microscopy.

The Young-Laplace and the force balance equations in this paper were derived from first principles for describing the shape of a deformed gas-liquid meniscus of a bubble and the forces exerted on a spherical particle as it undergoes attachment to an air bubble and its subsequent detachment. These two equations were developed based on the minimisation of free energy for the gravitational potential and the interfacial surface energies; the models also include a Lagrange multiplier to satisfy the bubble volume constraint.

The deformation of the interface was able to be successfully accounted for in the solved Young-Laplace equation by determining the correct value of $\lambda$ from image analysis of the high-speed videos obtained. The experimental force exerted on the particle was described as a function of the particle central polar angle and correlated very well with the forces calculated from the model using the parameters determined from each of the high-speed video frames.

Overall our research data show that the contact angle does not remain constant throughout particle attachment and detachment. Our work also indicates that the bubble profile does not remain spherical as a particle undergoes detachment. The outcomes of this paper provide a useful framework
for analysing the bubble deformation, interaction force, and change in contact angle during the attachment and detachment of the approach and retraction of a particle using AFM type technique.
References


Chapter 5

Drag force measurements on a stationary sphere floating at a planar surface in a shear flow field
Abstract

Detachment of particles from the bubble surface plays an important role in the flotation process where hydrophobic particles and hydrophilic particles are separated from each other in an aerated aqueous pulp. The literature indicates that particle detachment from air bubble depends strongly on the hydrodynamic forces acting on the particle. This paper is concerned with measuring the magnitude of the hydrodynamic forces exerted on a stationary particle floating at the air-water interface under the influence of a well-defined shear flow field. A new experimental technique has been developed using a force cantilever positioned at the surface of a small channel with fully developed laminar flow operating at intermediate Reynolds numbers. Pure water flows past a partially submerged spherical silica particle cantilevered using a carefully calibrated spring wire, enabling calculation of the force exerted on the particle using Hooke’s law. The water velocity was carefully controlled and determined using particle tracking velocimetry with the particle displacement measured using a laser triangulation system as used in typical AFM equipment. The results are analysed, and a simple relationship as a function of the three-phase contact angle is developed for the drag force acting on a partially submerged particle in a shear flow field.

1 Introduction

In mineral flotation, the collection of particles by the bubbles in the aqueous aerated pulp involves three key steps: collision, attachment and detachment [1]. Flotation relies on differences in surface properties, either naturally occurring hydrophobicity or that induced by the addition of surfactants.

Hydrodynamic interactions are the governing mechanism of the collision efficiency. Interfacial surface forces, represented by the three-phase contact angle, determine the attachment efficiency. The stability of the formed bubble-particle aggregates is a function of both interfacial forces and the hydrodynamics of the system [1].
The motion of a particle fully submerged in a fluid is influenced by the Stokes’ drag force. For a sphere immersed in an unbounded fluid, this is described as:

\[
F = -6\pi\mu(R)(V - W)
\]  

(1)

where \( R \) is the particle radius, \( \mu \) is the liquid viscosity, and \( V \) and \( W \) are the vectors of the particle and fluid velocities, respectively. When the particle approaches the bubble surface with an intervening liquid film, the hydrodynamic resistance increases rapidly. This deviation of the hydrodynamic resistance is due to the liquid film and can be accounted for using the hydrodynamic resistance functions [2]. For the case of interest in this paper, when the particle is attached to the air-water surface the hydrodynamic resistance functions will change again. The Stokes drag force Eq. (40) is modified to give

In the (radial) direction of the bubble-particle centreline:

\[
F_r = -6\pi\mu RV, f_1 + 6\pi\mu RW, f_2
\]  

(2)

In the (tangential) direction perpendicular to the centreline:

\[
F_t = -6\pi\mu RV, f_3 + 6\pi\mu RW, f_4
\]  

(3)

where the subscripts ‘r’ and ‘t’ describe the radial and tangential components of the drag force and the particle and fluid velocities, respectively, as shown in Figure 1. In this paper, we are concerned with the effects of the tangential velocity component of the fluid flow on a stationary particle. In a future paper, we will discuss the radial velocity component or stagnation flow field.

**Figure 1.** Schematic of a stationary spherical particle attached to an air-water interface in shear flow parallel to a slip planar gas-liquid interface and the local cylindrical coordinates \((\omega, \varphi, z)\). \( R \) is the particle radius, the central polar angle describing the contact point on the particle surface is given by \( \alpha \), and is equal to the three-phase contact angle, \( \theta \).
Research into the interaction between a solid particle approaching a solid surface has been well established with all four hydrodynamic resistance functions available as a function of the separation distance between the particle and the solid surface [2–4]. The drag force correction factor ($f_4$) for the slow viscous motion on a spherical particle just touching an immobile plane wall under shear flow has been calculated at $f_4 = 1.7009$ [3,5]. Here it should be noted, that the mobile liquid-air interface significantly differs from the solid-liquid interface in the magnitude of the liquid velocity tangential to the interface. In the immobile case, the velocity is zero (no slip), but it is different from zero in the case of the mobile liquid-air interface (slip).

For the interaction between a solid particle approaching an air bubble the hydrodynamic resistance functions are available as a function of the separation distance: $f_1$, $f_2$ [6], $f_3$ [7] and $f_4$ [8]. Of particular interest is the drag force correction for the case of a sphere sliding across the surface of an air bubble. A parabolic shear velocity profile was numerically solved, arriving at an asymptotic solution for the case where the sphere is just touching the surface (where $f_4 \rightarrow 1.2206$).

\[ \text{Figure 2. Comparison of experimental and theoretical drag force coefficients for particle motion correction.} \]

Reviewing the literature dealing with the case for when a spherical particle is attached to an interface, the focus has been on studying the motion of particles in a stationary flow field. Thus, the drag force correction arising from the particle motion, $f_3$, has been well researched in a variety of experimental and theoretical studies [9–16]. Some of the key results from these studies in determining the correction factor $f_3$ are shown in Figure 2.

These experimental studies typically involved using capillary or magnetic forces to force a floating particle to move. In one experiment, Petkov [17] manipulated a Teflon barrier to control the curvature of the fluid meniscus to create particle motion across the interface. Measurements of diffusion coefficients using three-dimensional real-time tracking of nano-sized particles attached to
an oil-water interface have also been undertaken [18,19]. Du, Liddle, and Berglund [18] found that there was a nonlinear relationship with the particle radius to the drag force suggesting the importance the contact angle has on the drag force. Using the modified Stokes law drag coefficient Dörr et al. [15] developed a numerical model for particles moving along an interface based on a vanishing viscosity ratio between the two phases. The model was devised specifically for contact angles of 90°, and the range extended using perturbation expansions to develop predicted values over the full range of 0° to 180°.

In this paper, we have developed a new experimental procedure to obtain the correction factor $f_4$ by investigating the shear flow force exerted on a stationary particle attached to an air-water interface, with different wetting (contact angle) properties. We use a custom built laser triangulation system, similar to the method that is used for operating the Atomic Force Microscopy (AFM) [20]. Using a laser and mounting the spherical particle on a carefully calibrated length of spring wire, we have directly measured the force exerted on the cantilevered particle at an air-water interface under a well-defined parabolic shear flow velocity profile.

2 Experimental Methods

2.1 Materials

The particles used to measure the force exerted by the water flow were spherical borosilicate glass beads (Sigma-Aldrich, Australia) with a diameter of approximately 2 mm and 5 mm. In the experiments, both hydrophilic and hydrophobic silica particles were used. The hydrophilic silica particles were cleaned ultrasonically in an alkaline cleaning solution prepared with potassium hydroxide, water, and ethanol (12.5:16:80 mass ratio) and rinsed several times with deionized (DI) water. The particles were stored in DI water to maintain the hydrophilic surface. Additional silica particles were hydrophobised by esterification in 1-octanol [21–24] to obtain equilibrium contact angles on a flat surface of around 85°. The hydrophobic silica particles were cleaned ultrasonically in ethanol and water then dried and stored in a desiccator. The silica particle was glued onto a thin wire (190 µm) using a small amount of epoxy resin (Selleys Araldite Super Strength, Australia). To determine the undisturbed water velocity profile polyamide seeding particles with a mean diameter of 50 µm (Dantec Dynamics, France) were used. The water used in all the experiments was freshly purified using a reverse osmosis RIO’s unit and an Ultrapure Academic Milli-Q system (Millipore, USA). The surface tension of the water was taken as 0.072 N/m, and this value was adopted for use in the calculations.
2.2 Experiment

The aim of the experiment was to accurately measure the force exerted on both a 5 mm and a 2 mm diameter silica particle attached to a free air-water interface as a function of water velocity and contact angle. Measurement of the force exerted on the particle was achieved by using a cantilever constructed from a short length of fine spring wire with a measurable spring constant. The air-water interface used in the experiments was the surface of an open flow channel operating under laminar conditions with DI water. To measure the force exerted by the water flow on the particle, the water flow rate was set, and the volumetric flow rate and the velocity profile below the surface were both measured. The silica particle was positioned at the water surface, measuring the submerged portion of the particle below the water surface to calculate the contact angle. The displacement of the particle downstream of the channel was measured, using a laser triangulation technique. The experimental arrangement is shown in Figure 3. In the experiments the water interface attached to the particle was meticulously controlled to be flat, meaning that the angle of inclination of the meniscus was equal to zero. For this scenario in can be shown that three phase contact angle is equal to the central polar angle [1].

Figure 3. Experimental arrangement showing a cross section of the laminar flow channel, high-speed camera, support structure, background lighting and the cantilevered particle.
Figure 4. Laminar water channel experimental system, not to scale, consisting of a head tank, control valve, laminar flow channel, sump and a centrifugal pump.

The arrangement of the water flow system is shown in Figure 4 and comprises of an 1 m long laminar flow channel (clear acrylic), 25.3 mm wide and 38 mm high, and a centrifugal pump (AP1050 Aquapro, China) to circulate the water through the system. The pump inlet from the sump box was specially designed not to draw in bubbles that would be broken up creating nano-bubbles throughout the system. The head tank, inflow, overflow and sump boxes were constructed from HDPE, with the system connected using round 20 mm PVC pipe. The total fluid volume of the flow system was approximately 8 L of DI water. The volumetric flow rate through the laminar flow channel was controlled using a brass gate valve; the isolation valves were PVC ball valves.
Due to the nature of the live load on the anti-vibration table, care had to be taken to minimize the vibrations created by the water circulation. The measures taken include a special weight for the flow channel and a carefully designed weir and overflow box to eliminate rippling back along the surface of the water in the channel. To ensure straight flow along the length of the channel a laminar flow device was positioned at the start of the channel, constructed from of a 50 mm length of filtration sponge. A constant head tank configuration was used to ensure no vibrations from the centrifugal pump were transferred to the flow profile in the channel. A photograph of the experimental rig is given in Figure 5.

To measure the volumetric flow rate of water through the laminar flow channel timed collection tests were undertaken, using a measuring cylinder and a stop watch, with the average of three measurements used as the result. To calculate the average water velocity and Reynolds number of the channel the water level was measured using a digital Vernier calliper (Maxwell, ME1002, China) at the location of the cantilevered particle. The water level was kept consistent along the length of the channel, achieved by introducing a slight slope to the channel.

To determine the undisturbed velocity profile just below the surface, particle tracking velocimetry was used with 50 µm polyamide seeding particles and a high-speed camera to track the
progress of the tracer particles (Figure 3). The arrangement comprises of a high-speed microscope camera video mounted on to laboratory jack, a PC for collecting the videos, a one meter long laminar open flow channel (clear acrylic), and a spherical silica test particle cantilevered to an x-y-z clamp and positioned in the centre of the channel. The laboratory jack is used to keep the camera level with the water surface as the water height changes proportionally with the volumetric flow rate.

As the high-speed camera microscope system requires back lighting, we need to ensure the water velocity measured is in the centre of the channel. To achieve this, the cantilevered particle is first submerged below the surface of the water, and the high-speed camera is focused on the edges of the submerged particle. The silica particle is raised from the channel, and a small quantity of polyamide particles is introduced into the channel inflow box, the high-speed camera records the progress of the polyamide particles at 2 000 frames per second through the region of interest. The video is analysed using a custom written MATLAB code developed for calculating the velocity of individual tracer particles. It is important to note that only the polyamide particles that are clearly in focus, identified using human eye, are analysed. This method allows us to capture the undisturbed velocity profile in the centre of the channel where the silica particle will be located.

Once a suitable tracer particle is identified, the MATLAB code analyses each frame of the video to determine the tracer particle diameter, the distance from the air-water interface and the change in location of the tracer particle centre from the previous frame. This is achieved using the built in Canny edge detection method supported by the edge function in MATLAB’s Image Analysis Toolbox, to generate a binary image of the edge profiles. The centre point and diameter of the tracer particle was determined by fitting the profile to an equation of a circle. The velocity was determined by the camera frame rate, the arc length of the tracer particle path and by using the ratio of pixels to length in µm, based on previous calibration images captured using the same lenses focal arrangement.

To measure the force exerted on the particle attached to the air-water interface a short length of spring wire was carefully calibrated and the spring constant determined using Hooke’s law, Eq. (4). To measure the displacement downstream of the silica particle, δz, a laser beam deflection system was setup as illustrated in Figure 6.

\[ F = k_c \delta z \]  

In the arrangement shown in Figure 6 a laser pointer (Class 2 laser output power of 1mW and 650 nm wavelength) is mounted using an optical clamp on a vertical and horizontal translation stage. To measure the distance from the cantilever to the wall, X, a laser distance measurer (Ryobi, RLM30, China) with an accuracy of ±2 mm was used. The length of the cantilever, l, was measured using the digital Vernier calliper along with the displacement of the laser spot, ∆S, from the zero flow position to the fully established flow rate. In the experiments, the distance X was around 2 500 mm, and the
vertical displacement of the laser point $\Delta S$ was between 1 and 20 mm. The ratio of the two lengths means that the angle of displacement, $\omega$, is only a fraction of a degree.

Figure 6. Schematic of laser beam triangulation system showing the silica particle attached to a length of spring wire, a small (2 mm) mirror, the laser diode and the portable clamp to hold the cantilever in place.

The cantilever deflection, $\delta z$, can then be calculated using Eq. (5) when the cantilever length, the distance between the cantilever and the wall, and the distance between the initial and final positions of the laser point on the wall are known.

$$\delta z = \frac{l}{2X} \Delta S$$  \hspace{1cm} (5)

Using the principle of similar triangles, Eq. (5) self corrects to account for the difference between the mirror placement and the centre of the particle. The correction is achieved by defining $l$ as the total spring wire length plus the particle radius, this allows for accurate calculation for the displacement particle centre.
Figure 7. Curve used to calculate the spring constant (75.566 µN/mm) for calibration of the 5 mm particle. Based on six data points and setting the intercept to zero.

Calibration of the cantilever was achieved by repositioning the clamp to hold the wire horizontally and using a set of three calibration weights applied to the tip of the wire. The displacement of the particle was performed using the same arrangement as shown in Figure 6, where instead of using the wall the ceiling of the laboratory was used. The measurement procedure was repeated twice and the results of Eqn. (5) are plotted in Figure 7, where the spring constant is determined using the equation of best fit.

When the cantilevered particle was positioned at the air-water interface, using the high-speed camera, a single image was captured showing the submerged proportion of the particle below the interface. The image was later analysed using another custom MATLAB code to determine the central polar angle, $\alpha$, describing the position of the interface on the particle surface. When the interface is flat this angle is equal to the contact angle, $\theta$ (see Figure ). Using the built in Canny edge detection method, a binary image of the edge profiles was generated. The image was rotated to align the interface in the direction of gravity; the angle rotation was determined by fitting a first order polynomial to the interface and calculating the angle of inclination. After the image rotation, the profile of the silica particle was determined by fitting the image data to an equation of a circle. The intercept of the air-water interface equation with the particle surface equation was determined and the polar angle calculated. The diameter of the particle was also calculated as a check using the ratio of pixels to length in µm, determined from previous calibration images captured using the same lenses focal arrangement used in the current experiments.
2.3 Computational methods

The simulation methodology to model the flow hydrodynamics of the experimental system is described in this section. The purpose of the modelling is to validate the use of the parabolic flow profile and provide a check to the velocity profile obtained using particle tracking velocimetry. The geometry of the simulated system is a simple rectangle, 600 mm long and 25.3 mm wide. The length represents the distance from the channel inlet to the position of the particle, the height of the geometry is varied for each case run as the water level changes with a change in the volumetric flow rate. Both the length and width measurements are the same geometry of the experimental system previously reported. The laminar flow in the column is governed by the Navier-Stokes equations, with no-slip boundary conditions on the column wall. In the model, only the water phase flow model with a laminar viscous model to solve the conservation of mass and momentum equations using a finite-volume based commercial CFD software package ANSYS Fluent (ANSYS Inc, v17.0, USA) with no CFD modelling of the particle is used. The computational domain was discretised in 410 832 control volumes and the Simple scheme was used for the pressure-velocity coupling. A first-order implicit formulation was used for the transient simulation. A steady state simulation was used with a value of $10^{-6}$ selected for both continuity and momentum convergence criteria. The discretization scheme chosen was least squares cell based for the gradient, and second order for the pressure, second order upwind for the momentum to assist in solving the predicted flow.

3 Results and Discussion

In developing the experimental procedure, initially, it was thought that hydrophilic particles should be used for the experiments to achieve a flat interface. Using hydrophilic particles works well for contact angles over a narrow region between 60° and 80°. To extend the range to between 40° and 130° hydrophobic particles were found to be well suited. By making use of the pinning and de-pinning effect discussed previously [25] and by lowering the particle to the point where the interface begins to slide and then raising the particle, a flat interface was able to be achieved for each measurement over the entire range reported.

The following section details the results obtained for two spherical hydrophobic particles with a diameter of 5 322 µm and 2 464 µm. For each particle size five different volumetric flow rates were tested, and while maintaining a constant flow rate through the channel, the position of the particle was changed measuring the force exerted on the particle over a range of contact angles.

3.1 Water velocity

During the experiments the volumetric flow rate and the water level were recorded and are detailed in Table 1 and Table 2 for the 5 and 2 mm particles, respectively. The flow regime in the
open channel can be defined by the Reynolds number and has been calculated using the hydraulic
diameter of the channel as the characteristic length and the average channel velocity. The results of
the calculation indicate that the water flow in the channel is clearly classified as laminar for all the
experimental test cases. The water density and viscosity have both been corrected for the actual fluid
temperature measured during the experiments.

**Table 1.** Reynolds number as a function of the volumetric flow rate using both the hydraulic diameter
and the particle diameter for the 5 mm particle.

<table>
<thead>
<tr>
<th>Volumetric flow rate</th>
<th>Water height</th>
<th>Average velocity</th>
<th>Reynolds number Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td>mL/s</td>
<td>mm</td>
<td>cm/s</td>
<td></td>
</tr>
<tr>
<td>7.3</td>
<td>16.75</td>
<td>1.7</td>
<td>504</td>
</tr>
<tr>
<td>11.9</td>
<td>17.76</td>
<td>2.6</td>
<td>773</td>
</tr>
<tr>
<td>16.6</td>
<td>19.39</td>
<td>3.3</td>
<td>1 049</td>
</tr>
<tr>
<td>18.8</td>
<td>19.78</td>
<td>3.7</td>
<td>1 174</td>
</tr>
<tr>
<td>20.7</td>
<td>20.91</td>
<td>3.9</td>
<td>1 224</td>
</tr>
</tbody>
</table>

**Table 2.** Reynolds number as a function of the volumetric flow rate using both the hydraulic diameter
and the particle diameter for the 2 mm particle.

<table>
<thead>
<tr>
<th>Volumetric flow rate</th>
<th>Water height</th>
<th>Average velocity</th>
<th>Reynolds number Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td>mL/s</td>
<td>mm</td>
<td>cm/s</td>
<td></td>
</tr>
<tr>
<td>16.1</td>
<td>16.81</td>
<td>3.7</td>
<td>1 145</td>
</tr>
<tr>
<td>17.3</td>
<td>16.82</td>
<td>4.0</td>
<td>1 220</td>
</tr>
<tr>
<td>19.1</td>
<td>16.83</td>
<td>4.4</td>
<td>1 345</td>
</tr>
<tr>
<td>19.7</td>
<td>17.36</td>
<td>4.4</td>
<td>1 397</td>
</tr>
<tr>
<td>25.3</td>
<td>18.16</td>
<td>5.4</td>
<td>1 707</td>
</tr>
</tbody>
</table>

To determine the unperturbed velocity profile below the water surface, the high-speed camera
was used to record the progress of polyamide seeding particles. The water velocity could be
calculated using the camera frame rate and the distance travelled by the tracer particle through the
field of focus. The velocity profile was also modelled using CFD based on the experimental
volumetric flow rate. The results from the two methodologies are plotted in Figure 8 for the 5 mm
particle and Figure 9 for the 2 mm particle. For each velocity profile, between 20-30 tracer particles
were selected to be tracked using the Matlab code described previously, and the calculated velocities
are plotted as a function of distance below the air-water interface. The vertical velocity profile is
taken in the centre of channel at a point 600 mm from the inlet. The full velocity profile cross section for a single case is shown in Figure 10.

**Figure 8.** Velocity as a function of the position below the water surface for the 5 mm particle. The experimental points are obtained from the tracer particles and the line results from the CFD model.

**Figure 9.** Velocity as a function of the position below the water surface for the 2 mm particle. The experimental points are obtained from the tracer particles and the line results from the CFD model.
Figure 10. Velocity contour for the channel 600 mm from the water inlet. The results are for the highest flow rate for the 5 mm particle with a water height of 20.9 mm with an average velocity at the inlet of 3.9 cm/s.

Analysing the velocities reported in Figure 8 and Figure 9, indicate that the results from the high-speed camera are in agreement with the CFD modelling results. The parabolic velocity profile generated by the CFD models also compares well with the fundamental principles of open channel flow [26–28] where the flow profile has been shown to be parabolic with the maximum velocity occurring just below the water surface and reducing to zero at the bottom of the channel.

3.2 Measured force

Force measurements for the 5 mm particle are plotted in Figure 11 as a function of the three-phase contact angle.
Figure 11. Measured force for the 5 mm particle as a function of the position of the central polar angle, $\alpha$, on the particle surface at the five different volumetric flow rates.

Analysing the data in Figure 11, we can fit an exponential curve to each constant volumetric flow rate data set listed in Table 3. Using equations in Table 3, we can determine a value for the force when the interface is pinned on the particle surface at a polar angle of 90°. When the results are plotted as a function of the volumetric flow rates we obtain Figure 12.

Table 3. Equations of best fit for the measured force for the 5 mm particle.

<table>
<thead>
<tr>
<th>Volumetric flow rate</th>
<th>Force equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>mL/s</td>
<td>$\mu N$</td>
</tr>
<tr>
<td>7.3</td>
<td>$F = 12.901e^{-0.014\alpha}$</td>
</tr>
<tr>
<td>11.9</td>
<td>$F = 91.474e^{-0.033\alpha}$</td>
</tr>
<tr>
<td>16.6</td>
<td>$F = 55.949e^{-0.024\alpha}$</td>
</tr>
<tr>
<td>18.8</td>
<td>$F = 23.313e^{-0.012\alpha}$</td>
</tr>
<tr>
<td>20.7</td>
<td>$F = 25.535e^{-0.012\alpha}$</td>
</tr>
</tbody>
</table>

Similarly, for the 2 mm particle, we can generate equations of best fit for the measured force (Table 4), and a plot of the drag force as a function of the measured volumetric flow rate (Figure 14). The results shown in Figure 12 and Figure 14 provides a simple method to visualise the increase in the measured force exerted on the particle as the volumetric flow rate increases.
Figure 12. Correlation of the drag force exerted on the 5 mm particle submerged with a polar angle of 90° calculated from the equations of best fit from the experimental data, (listed in Table 3), as a function of the measured volumetric flow rate.

Figure 13. Measured force for 2 mm particle as a function of the position of the central polar angle, $\alpha$, on the particle surface for five different volumetric flow rates.

Table 4. Equations of best fit for the measured force for the 2 mm particle.

<table>
<thead>
<tr>
<th>Volumetric flow rate</th>
<th>Force equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>mL/s</td>
<td>$\mu$N</td>
</tr>
<tr>
<td>16.1</td>
<td>$F = 30.770e^{-0.025\alpha}$</td>
</tr>
<tr>
<td>17.3</td>
<td>$F = 31.848e^{-0.025\alpha}$</td>
</tr>
<tr>
<td>19.1</td>
<td>$F = 35.084e^{-0.026\alpha}$</td>
</tr>
<tr>
<td>19.7</td>
<td>$F = 18.089e^{-0.017\alpha}$</td>
</tr>
<tr>
<td>25.3</td>
<td>$F = 30.678e^{-0.022\alpha}$</td>
</tr>
</tbody>
</table>
**Figure 14.** Correlation of the drag force exerted on the 2 mm particle submerged with a polar angle of 90° calculated from the equations of best fit from the experimental data (Table 4), against the measured volumetric flow rate.

### 3.3 Drag force correction factor

In the literature, the velocity used to calculate Stokes drag force correction factors is based on the unperturbed velocity acting on the particle centre at infinity. This presents a problem with contact angles greater than 90° as the centre of the particle is above the water surface. To overcome this problem, the centre of mass of the immersed portion of the particle was used as the location to determine the unperturbed velocity, represented by:

\[
\bar{c} = 3R \frac{\cos^4 (\alpha/2)}{2 + \cos(\alpha)}
\]

Where \( R \) is the particle radius, \( \alpha \) is the central polar angle describing the position of the contact line, and \( \bar{c} \) is the geometric centre of the portion of the particle below the water surface. We can then use the results of the CFD modelling for each case as shown in Figure 8 and Figure 9 to determine the velocity acting on the centroid of the submerged portion of the silica particle.

As the particle is stationary with no rotation, we can re-write Eq. (42), solving for the drag force correction factor due to the water velocity, giving:

\[
f_d = \frac{F_t}{6\pi\mu RW_t}
\]

Using the experimental data to solve Eq. (7) we can determine the drag force correction factor for the tangential water velocity as a function of the central polar angle on the particle surface. These data are given in Figure 15 and Figure 16 for the 5 mm and 2 mm particles, respectively.
Comparing the correction factors presented in Figure 15 and Figure 16 we can see the results are highly comparable given the difference in particle size. If we also consider the literature values shown in Figure 20 and the work of Nguyen and Jameson [8] and Brenner [29] it is possible to say that the correction factor $f_4$ will approach a finite maximum as $\alpha$ approaches zero. For the limit when $\alpha$ approaches $180^\circ$, $f_4$ will approach zero. Based on these limits we can apply the following equation to model the experimental data.
\[ f_{d(model)} = \frac{K}{4} (2 + 3\cos \alpha - \cos^3 \alpha) \] (8)

Using the experimental data shown in Figure 15 and Figure 16 the value for the model fitting parameter \( K \) in Eq. (8), was found to equal 4.701 by minimising the least squares error. The plot of the modelled data fit is shown in Figure 17 as a function of the position of the central polar angle, \( \alpha \), on the particle surface.

\[ \text{Figure 17. Comparison of the correction factor and contact angle for the 5 mm and the 2 mm particles. Modelled fit data is also shown.} \]

3.5 **Effect of curved interface**

In developing the experimental procedure to achieve a flat-water interface at the three-phase contact point, it was initially thought that hydrophilic particles should be used. The use of hydrophilic particles worked well for obtaining contact angles between 60° to 80°, when the particle was mostly immersed. But attempting to lift the particle to achieve polar positions greater than 80°, resulted in a curved interface with the three-phase contact remaining attached around 80-85°. This means that for the correction factors in reported in Figure 18 over the range of 80° to 130° the air water interface is not flat but curves upwards to meet the particle surface.
Figure 18. Results initially obtained using a 2 mm hydrophilic particle. Illustrating the effect due to the presence of a meniscus on the particle surface for polar positions with an angle greater than 80°. Note that for the model, the central polar angle is equal to the contact angle but this is not true for the experimental data as the meniscus is inclined.

We were unable to quantify the angle of inclination of the meniscus on the particle, due to the presence of the meniscus on the channel walls. The presence of the meniscus on the channel wall also means that the angles reported in Figure 18 are not where the interface is pinned on the particle but where the flat interface at a short distance from the particle intersects with the particle. To remove the shadow created by the meniscus on the channel walls hydrophobisation of the acrylic surface was carried out using a trace amount of petroleum jelly. This removed the meniscus on the channel wall allowing for visualising the precise location of the pinned interface on the particle surface, helping to manipulate the position of the particle to have the flat-water interface.

We have decided to include Figure 18 as it highlights the effect brought about by the presence of a curved meniscus on the particle and how it significantly increases the drag force correction factor. Also, it should be considered that the meniscus has a downwards force, meaning that the drag force correction factors in Figure 18 are under reported.

4 Conclusions

In this paper, the drag force of both 2 and 5 mm spherical particles attached to an air-water interface was measured using a specially designed open laminar flow channel. The water velocity profile was obtained by using high-speed video microscopy to capture the progress of tracer particles in the channel. The velocities obtained using the high-speed camera were shown to agree with CFD modelling based on experimental measurements. The drag force exerted on a stationary particle was directly measured using a carefully calibrated spring wire. A clear correlation between the drag force correction factor, $f_i$, and the contact angle can be seen in the results obtained.
References


Chapter 6

Drag force measurements on a stationary sphere floating at a planar surface in a stagnation flow field
Abstract

Detachment of particles from the bubble surface plays an important role in the flotation process where hydrophobic particles and hydrophilic particles are separated from each other in an aerated aqueous pulp. The literature indicates that particle detachment from air bubbles depends strongly on the hydrodynamic conditions acting on the particle. This paper is concerned with the magnitude of the hydrodynamic forces exerted on a stationary particle floating at the air-water interface in a well-defined stagnation flow field. A new experimental technique has been developed using a small vertical column with fully developed laminar flow operating at low Reynolds numbers. Pure water flows past a partially submerged spherical silica particle suspended from a force balance directly measuring the exerted flow force on the particle. The results are analysed and a simple relationship is proposed as a function of the three-phase contact angle for calculating the correction factor to the modified Stokes drag force equation when a stagnation flow field is acting on a particle floating at a flat air-water interface.

1 Introduction

Interactions between bubbles and particles in aqueous solutions plays an important role in various industrial processes, particularly in froth flotation, the technique widely used for the separation of mineral particles, treatment of wastewater and the recycling of paper fibres [1]. Since the selective attachment of air bubbles to target particles determines the separation efficiency of hydrophobic and hydrophilic particles in a flotation cell, understanding the hydrodynamic bubble–particle interactions in froth flotation is critical [2,3]. An important feature of bubble–particle interaction is the influence of hydrodynamic forces on particles attached to a free air-water interface.

When a particle is attached to an interface, corrections have to be made to Stokes’ drag force equation, given by Eq. (40), for a sphere immersed in an unbounded fluid:

\[ \mathbf{F} = -6\pi\mu R (\mathbf{V} - \mathbf{W}) \]

(1)

Where \( R \) is the particle radius, \( \mu \) is the liquid viscosity, and \( \mathbf{V} \) and \( \mathbf{W} \) are the vectors of the particle and fluid velocities, respectively. When the particle is attached to an air-water interface, the
hydrodynamic resistances change. This deviation to the hydrodynamic resistance can be accounted for using the hydrodynamic resistance functions [4]. For the case of interest, when the particle is attached to the air-water surface Stokes drag force Eq. (40) is modified to give

In the (radial) direction of the bubble-particle centreline:

$$ F_r = -6\pi \mu R V f_1 + 6\pi \mu R W f_2 $$

(2)

In the (tangential) direction perpendicular to the centreline:

$$ F_t = -6\pi \mu R V f_3 + 6\pi \mu R W f_4 $$

(3)

where the subscripts ‘r’ and ‘t’ describe the radial and tangential components of the drag force and the particle and fluid velocities, respectively, as per Figure 1. In this paper, we are concerned with the effects of the radial velocity component of the fluid flow. In a previous paper, we discussed the tangential velocity component or shear flow field on the drag force.

![Figure 1](image)

**Figure 1.** Schematic of a stationary spherical particle attached to an air-water interface in shear flow parallel to a slip planar air-water interface and the local azimuthal cylindrical coordinates ($\omega$, $z$). Where $R$ is the particle radius, the central polar angle describing the contact point on the particle surface is given by $\alpha$, and is equal to the three-phase contact angle, $\theta$.

Reviewing the literature studying the hydrodynamic interactions for a spherical particle attached to an interface, the research focus has been on understanding the motion of the particles in stationary flow fields. Thus, the drag force correction arising from the particle motion, $f_3$, has been well researched in a variety of experimental and theoretical studies having been undertaken [5–12].

The subject of a spherical particle straddling a gas–liquid interface relevant to the stability of particle–bubble capture with a defined contact angle in an extensional flow has been numerically studied by Stoos and Leal [13]. The problem addressed in their work was the case where an attached particle is swept to the rear stagnation point of a bubble and is exposed to an axisymmetric straining
flow tending to detach the particle from the interface. Stoos and Leal [13] adopted numerical treatment to the problem using a boundary-integral technique to consider a spherical particle contacting a free deformable interface in the presence of a steady uniaxial extensional flow, presenting the results as a set of dimensionless critical capillary numbers for system stability.

![Graph showing critical capillary number for particle detachment as a function of contact angle](image)

**Figure 2.** Critical capillary number for particle detachment as a function of contact angle [13]. Where the capillary number is defined by $Ca = \mu GR / \sigma$ and $Cg = \mu G / Rg$, $G$ is the shear stress, $R$ is the particle radius, $\mu$ is the liquid viscosity and $\sigma$ is the surface tension.

The results shown in Figure 2 describe for any given capillary system the critical capillary number beyond which the system would be unstable and the attached particle would detach from the interface by the straining flow pressure force. The results were obtained at by calculating a set of equilibrium conditions where the buoyancy, capillary and hydrodynamic forces are balanced, and preceding to manually adjust the values for $Ca$ and $Cg$ until an unstable configuration was reached. The results for the case where the contact angle is 90° provides a valuable reference for this experimental study as the interface is relatively flat and is comparable to this work. The other two cases in Figure 2 are not comparable as when the contact angle was 160°, the interface has significant deformation, and when the contact angle was 20°, the authors were unable to calculate a stable configuration using their estimates for $Ca$ and $Cg$.

Another useful reference can be obtained, when considering the case where a particle is approaching a mobile bubble surface while subjected to a stagnation flow field. The drag force correction factor to Eq. (2) has been predicted using a tangent-sphere coordinate system and was found to equal $f_2 = 2.039$ [14]. This value represents the limit as the particle is just touching the bubble surface and is also close to the previously calculated value in the literature of 2.034 [15].

This paper presents the results of work conducted using a new experimental technique to measure the drag force on a spherical particle floating at the air-water interface in a well-defined
stagnation flow field. The measurements were made using a modified sphere tensiometry apparatus to directly measure the force exerted on the floating particle. The stagnation flow field was generated using a specially designed flow cell, operating at low Reynold’s numbers with pure water flowing vertically upwards and overflowing the column walls.

2 Experimental Methods

2.1 Materials

The particles used were two hydrophobic spherical borosilicate glass beads (Sigma-Aldrich, Australia) with a diameter of 3.9 and 4.6 millimeters. To prepare the silica particles used in the experiments they were first cleaned ultrasonically in alkaline cleaning solutions prepared from potassium hydroxide, water, and ethanol and (12.5:16:80 mass ratio) and vigorously rinsed many times with deionized (DI) water. The silica particles were then hydrophobized by esterification in 1-octanol [16–19] to obtain contact angles of around 85°. The hydrophobic silica particles were cleaned ultrasonically in ethanol and water then dried and stored in a desiccator. The silica particle was glued onto a thin wire (500 μm) using a small amount of epoxy resin (Selleys Araldite Super Strength, Australia). The water used in all the experiments was freshly purified using a reverse osmosis RIO’s unit and an Ultrapure Academic Milli-Q system (Millipore, USA). The surface tension of the water was taken as 0.072 N/m, and this value was adopted for use in all calculations.

2.2 Experiment

The aim of the experiment was to be able to accurately measure the force exerted on two different silica particles attached to a free air-water interface as a function of water velocity and wetting properties e.g. the three-phase contact angle. The experimental setup comprised a force balance (XS205 Dual Range Analytical Balance with 0.01 mg accuracy, Mettler Toledo, USA), a motorized laboratory jack (L490MZ/M Motorized Lab Jack, Thorlabs, Cambridgeshire, UK), a custom built vertical laminar flow cell (glass), a spherical test particle mounted to the bottom of the force sensor, a high-speed microscope camera video (Photron Fastcam SA1.1, USA), and a PC for synchronizing the motion of the motorized stage and recording the force measurement data.

The vertical laminar flow cell was custom designed and constructed from blown glass, the inner flow tube had a constant diameter of 36.3 mm with a 90 mm clearance above a flow straightener (20 mm thick filtration sponge) the outer cylinder diameter for capturing the overflow was 55 mm. The experimental setup, including the arrangement of the circulation system to control the water flow through the vertical laminar flow cell is shown in Figure 3, a photo of the actual setup can be seen in Figure 4.
A centrifugal pump (DC40, China) was used to circulate the water through the system. The pump inlet from the sump box and the outlet of the head were specially designed not to draw in bubbles that could be broken up creating nano-bubbles throughout the system. The head tank, inflow, overflow and sump boxes were constructed from HDPE, with the system connected using round 12 mm clear vinyl tubing. The total fluid volume of the flow system was approximately 2 L (of DI water). The volumetric flow rate through the laminar flow channel was controlled using a built-in control valve on a calibrated rotameter (EW-32461-40, Cole-Palmer, US).

**Figure 3.** Experimental setup, not to scale, consisting of a force balance, a motorized lab jack, a laminar flow cell, suspended test particle, a high-speed camera with background lighting, a PC for synchronizing and data collection, and the water circulation system.
Figure 4. Photograph of the experimental rig. 1. Laminar flow cell. 2. High-speed microscope camera. 3. Force balance. 4. Local airflow isolation box. 5. Motorized lab jack. 6. Rotameter with built-in control valve. 7. Pump and sump. 8. Head tank (out of sight).

The system was filled with Milli-Q water, placing the flow cell on the motorized lab jack. To minimize vibration and disturbances, the whole setup was placed on a vibration isolation table in an air-conditioned laboratory. The water temperature was kept constant at 20°C, based on this temperature the water density and viscosity used in the calculations was 998 kg/m³ and 1.001 cP respectively. The position of the particle relative to the flow cell was measured and carefully positioned to locate the particle centrally in the flow cell. The flow cell and the motorized stage were enclosed in a clear acrylic box to isolate the effect of any local airflow on the test particle and disturbances to the air-liquid interface.

To measure the force, the hydrophobic silica particle was suspended using a short length wire from the force balance and the balance zeroed. The water flow rate was set to the lowest flow rate reported, using the rotameter. Using the motorized lab jack the flow cell was raised up to the particle, attaching the silica particle to the water surface.

To control the wetting properties of the silica particles at different contact angles, use of the pinning and de-pinning effect discussed in previous work was adopted [20]. The interface was controlled to be flat by lowering the particle to the point where the interface began to slide and then by raising the particle, a flat interface was able to be achieved for each measurement over the entire range reported. An experimental photograph illustrating the flat-water surface achieved in the experiments is shown in Figure 5.
Figure 5. Image showing the 4.6 mm particle suspended from the force balance and attached to a flat-water surface under the influence of a stagnation flow field.

As the water interface is flat at the three-phase contact point, the vertical component of the capillary force can be ignored along with line tension effects, which may important for very small particles. When there is no angle of inclination of the meniscus it can be shown that three-phase contact angle is equal to the central polar angle [1]. As the force balance was zeroed with the suspended particle prior to partially submerging the particle, the buoyancy force needed to be corrected. This force is dependent on the volume of liquid displaced by the immersed portion of the particle in the water, which can be calculated using the following equation [21]:

$$F_b = \frac{\pi R^3 \delta g}{3} \left(2 + 3 \cos \alpha - \cos^3 \alpha\right)$$  \hspace{1cm} (4)

where $R$ is the particle radius, $\delta$ is the density of liquid, $g$ is the acceleration of gravity constant and $\alpha$ is the central angle measured from the centre of the particle between the surface tension direction and the upwards vertical.

When the particle was positioned at the interface an image of the portion of the particle above the water surface was captured using the high-speed camera. The image was later analysed using a custom MATLAB code to determine the central polar angle, $\alpha$, describing the position of the interface on the particle surface, which is equal to the contact angle, $\theta$. Using the built-in Canny edge detection method in MATLAB, a binary image of the edge profiles was generated. The image was rotated to align the interface in the direction of gravity; the angle rotation was determined by fitting a first order polynomial to the interface and calculating the angle of inclination. After the image rotation, the air-water interface position was recalculated and the profile of the silica particle was determined by fitting
the image data to an equation of a circle. The intercept of the air-water interface equation and the
equation describing the particle surface was found, and the polar angle calculated.

Both the force balance and laboratory jack were operated by Lab View software developed
in-house and installed on a PC to manipulate the vertical lab jack displacement (in the air flow
isolation box to minimize vibrations). The force was measured for a period of 20 s, taking the average
while ensuring there was a low standard deviation of the force measurement recorded for the time
period. The flowrate was increased to the next increment, another image of the top segment of the
particle was captured and the measured force recorded.

Calibration of the flowmeter was conducted using a series of timed collection volume tests
using a measuring cylinder and a stop watch, taking the average of three measurements. The results
are plotted in Figure 6.

![Figure 6](image.png)

**Figure 6.** Calibration curve of the flowmeter giving a calibration factor of 1.093 for DI water at 20°C.

### 2.3 Computational methods

The simulation methodology to model the flow hydrodynamics of the experimental system is
described in this section. The schematic diagram of the simulated system is depicted in Figure 7 and
is the same geometry of the experimental system previously reported. The laminar flow in the column
is governed by the Navier-Stokes equations, with no-slip boundary conditions on the column wall.
In the model, two phase flow equations were developed using an implicit volume of fluid (VOF)
model with a laminar viscous model to solve the conservation of mass and momentum equations
using a finite-volume based commercial CFD software package ANSYS Fluent (ANSYS Inc, v17.0,
USA). No CFD modelling of the particle has been undertaken in this work. The computational
domain was discretised in 60 192 control volumes. The Simple scheme was used for the pressure-
velocity coupling, with the second-order upwind discretization scheme. A first-order implicit
formulation was used for the transient simulation. A variable time-step was used for the simulations.
with a global Courant criteria of 0.5. A value of $10^{-4}$ was selected for both continuity and momentum convergence criteria. The discretization scheme chosen was least squares cell based for the gradient, body force weighted for the pressure, second order upwind for the momentum, and compressive for the volume fraction to assist in solving the predicted flow.

Figure 7. Sketch of the modelled geometry used to generate the mesh for modelling the water flow through the column.

3 Results and Discussion

3.1 Water velocity modelling

This section outline the inputs and the results obtained from the CFD simulation. The two phases selected to be modelled were air and water at 20°C, based on the experimental conditions. The modelling of the phases was completed using the volume of fluid (VOF) model with air set as the primary phase and water the secondary phase. To model the phase interactions, the surface tension between water and air was set to a constant value of 0.072 N/m. This created a few problems in the modelling, as the simulation would calculate the flow over the top of the column walls in drips (slug flow) instead of a constant stream as observed in the experiments. By carefully modifying the under-relaxation factors for the body force and momentum, depending on the flow rate, a constant flow over the channel walls was able to be modelled, as seen in Figure 8.

For the CFD model (the location of the input and output boundary conditions for the simulation are shown in Figure 7) the water flow would enter the simulation at the column inlet as a constant velocity, calculated from the experimental volumetric flow rate and column diameter, see...
Table 1. The water would fill the column and overflow running down the walls of the column and exiting through the outlet. The ambient boundary condition for the simulation was defined as a pressure outlet to account for air flow through the model created by the water motion.

Table 1. Flow rates used in the experiment, raw and corrected, and calculated average velocities (using the column diameter and the Reynolds number).

<table>
<thead>
<tr>
<th>Flowmeter reading</th>
<th>Corrected volumetric flow rate</th>
<th>Average velocity</th>
<th>Reynolds number</th>
</tr>
</thead>
<tbody>
<tr>
<td>mL/min</td>
<td>mL/min</td>
<td>cm/s</td>
<td></td>
</tr>
<tr>
<td>400</td>
<td>404</td>
<td>0.64</td>
<td>228</td>
</tr>
<tr>
<td>500</td>
<td>505</td>
<td>0.80</td>
<td>284</td>
</tr>
<tr>
<td>600</td>
<td>606</td>
<td>0.96</td>
<td>341</td>
</tr>
<tr>
<td>700</td>
<td>707</td>
<td>1.12</td>
<td>398</td>
</tr>
<tr>
<td>800</td>
<td>807</td>
<td>1.28</td>
<td>455</td>
</tr>
<tr>
<td>900</td>
<td>908</td>
<td>1.44</td>
<td>512</td>
</tr>
<tr>
<td>1,000</td>
<td>1,009</td>
<td>1.60</td>
<td>569</td>
</tr>
<tr>
<td>1,100</td>
<td>1,110</td>
<td>1.76</td>
<td>626</td>
</tr>
</tbody>
</table>

To simplify the simulation calculation requirements only a two-dimensional cross-section was modelled. As the flow is axisymmetric, only the flow in the central plane is required to be modelled.
Figure 8. CFD showing the volume fraction of the water phase in the simulation.

Velocity contours obtained from the CFD model for both the water and air phases is shown in Figure 9 for the maximum experimental flow rate, using an average velocity of 1.76 cm/s. While Figure 10 shows the vertical component of the water velocity, corresponding to the average velocity of 1.76 cm/s.

Figure 9. CFD results velocity contours including both water and air phases. Corresponding to the maximum experimental flow rate tested, with an average velocity of 1.76 cm/s.
Figure 10. CFD contours showing the vertical component of the velocity for the water phase only. Corresponding to the maximum experimental flow rate tested, with an average velocity of 1.76 cm/s.

To illustrate that the CFD results correspond to a stagnation flow field, Figure 11 is a plot of the axisymmetric velocity streamlines for the water phase only.

Figure 11. CFD results showing a close-up of the axisymmetric velocity streamlines for the water phase only. Corresponding to the maximum experimental flow rate tested, with an average velocity of 1.76 cm/s.

The velocity profiles for each of the experimental flow rates tested and tabulated in Table were obtained by running separate CFD models. The results for the y-component of the velocity
profile in the centre of the column are plotted in Figure 12 as a function of the distance below the interface.

![Figure 12](image)

**Figure 12.** CFD results for the y-component of the velocity profile in the centre of the column as a function of the distance below the interface. The velocity profiles shown are for all the experimental flow rates tested, the legend indicates the average velocity for each experimental flow rate.

3.2 **Measured force**

Force measurements for the 3.9 mm particle are plotted in Figure 13 as a function of the three-phase contact angle. The results shown are for four typical experimental runs, showing the measured force, calculated buoyancy force, and the adjusted force corrected for buoyancy. It is important to note that each of the experimental runs are for different contact angles. Additionally, as the water velocity increases the water level also slightly increases changing the contact point of the interface.
Figure 13. Four typical runs for the 3.9 mm particle showing the measured force, calculated buoyancy force, and the corrected force measurement accounting for buoyancy.

Figure 14. Average velocity as a function of the location of the three-phase contact line on the particle surface, $\alpha$. Measurements are for the 3.9 mm diameter particle, noting that the central polar angle is equal to the contact angle, $\theta$, as the interface is flat.
Using the same experimental runs as shown in Figure 13, the average velocity was plotted as a function of the contact angle in Figure 14. This is interesting as it shows the change of the contact angle due to the water level change by increasing the water flow rate.

![Figure 14](image)

**Figure 15.** Measured force, accounting for buoyancy, as a function of the location of the three-phase contact line described by the central polar angle in degrees.

The measured drag force exerted on each of the two particles is plotted in Figure 15 as a function of the three-phase contact angle for 15 separate experimental runs. It is important to note that the force measurements in Figure 15 do not take into account the magnitude of the water velocity. In addition, the limited number of data points and the range of contact angles shown is relatively narrow; this is due to the difficulty in maintaining a flat-water interface at the contact point on the particle as the water velocity changes.

### 3.3 Drag force correction factor

As the particle is stationary and non-rotating we can re-write Eq. (2) to solve for the drag force correction factor:

$$F_r = 6πμRW_r f_2$$

In Eq. (5), the reference velocity used to calculate the Stokes drag force correction factor is based on the velocity acting on the particle centre at infinity [15]. As the reference condition is laminar pipe flow we can use the simple relationship between the maximum and average velocity that is well understood in fluid dynamics [22,23].

$$W_r = 2W_{\text{average}}$$

(6)
Examining the velocity profiles obtained from the CFD simulation in Figure 12 we can see that the flow profile in the model is not fully developed, as the stagnation flow field alters the dynamics in the model.

**Table 2.** Comparison of the average velocity calculated from the volumetric flow rate to the maximum velocity in the centre of the flow cell as calculated by the CFD model and Eq. (6).

<table>
<thead>
<tr>
<th>Average velocity cm/s</th>
<th>Max velocity from CFD results cm/s</th>
<th>Max velocity from Eq. (6) cm/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.64</td>
<td>0.75</td>
<td>1.28</td>
</tr>
<tr>
<td>0.80</td>
<td>0.92</td>
<td>1.60</td>
</tr>
<tr>
<td>0.96</td>
<td>1.09</td>
<td>1.92</td>
</tr>
<tr>
<td>1.12</td>
<td>1.27</td>
<td>2.24</td>
</tr>
<tr>
<td>1.28</td>
<td>1.44</td>
<td>2.56</td>
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<td>1.44</td>
<td>1.61</td>
<td>2.88</td>
</tr>
<tr>
<td>1.60</td>
<td>1.78</td>
<td>3.20</td>
</tr>
<tr>
<td>1.76</td>
<td>1.94</td>
<td>3.52</td>
</tr>
</tbody>
</table>

Using the velocities obtained from Eq. (6) and tabulated in Table 2, we can determine the correction factor $f_2$ for the modified Stokes drag force. If we use the velocities predicted by the CFD model, we obtain correction factors in the region of 100-500 times the standard drag force.

**Figure 16.** Plot of the drag force correction factor as a function of the central polar angle, $\alpha$, in degrees.
3.4 General discussion

The correction factor values calculated from the experimental data are shown in Figure 16. These are comparable to the work of Dukhin and Rulev [15] along with Nguyen and Evans [14] where the value for $f_2$ was calculated to equal 2.039 for the case of an approaching particle to a bubble just touching the surface. Furthermore, it is possible to say that the correction factor $f_2$ will approach a finite maximum as $\alpha$ approaches zero. For the limit when $\alpha$ approaches $180^\circ$ $f_2$ should approach zero. Based on these assumed limits we can apply the following equation to model the experimental data.

$$f_{2(\text{model})} = \frac{K}{4}(2 + 3\cos \alpha - \cos^3 \alpha)$$

(7)

Based on the experimental data the fitting parameter for the constant $K$ in Eq. (7) was found to equal 3.421 by minimising the least squares error. The model is plotted with the experimental data in Figure 17.

![Figure 17](image)

**Figure 17.** Plot of the drag force correction factor as a function of the central polar angle, $\alpha$, calculated from the experimental data, and also showing the predicted model data based on Eq. (7).

To better judge the measure of fit for the model, Eqs. (5) and (7) were used to calculate the expected drag force for each contact angle measured. The results are plotted in Figure 18 as a function of the experimentally measured force.
Comparison of the experimental results in Figure 17 can be compared to the numerical results provided by Stoos and Leal [13] in Figure 2 when the contact angle is equal to 90°. The experimental data reported in this paper is over the range 70° to 110° and in Stoos and Leal work the interface is relatively flat when the contact angle is 90°. The other two values in Figure 2 for 20° and 160° are well outside the range of the experimental data collected. It is also worth noting that no stable configurations were calculated for the 20° case and the value for the critical capillary number is likely to be much smaller than what is reported in Figure 2. For the 160° case there was significant deformation of the air-water interface.

The strength of the particle attachment in an air-water interface can be quantified using a force balance of the adhesive forces to the detaching forces. Using a force balance we can predict the required hydrodynamic force to destabilize the capillary system. A simplification of the force balance has been condensed into Eq. (8) by Nguyen [24]. This equation incorporates an approximation of the adhesive forces of capillary, hydrostatic pressure and the buoyancy of the particle volume immersed in the liquid, along with the detaching force of the particle weight.

\[
T = \pi R \sigma \left(1 - \cos \theta\right) \left[1 + 0.016 \frac{R}{L}\right]
\]

(8)

Where \(T\) is the force of attachment holding the particle at the interface, \(L = \sqrt{\sigma / \delta g}\) is the capillary length, \(\theta\) is the three-phase contact angle, \(R\) is the particle radius, \(\delta\) is the water density, \(g\) is the acceleration due to gravity, and \(\sigma\) is the liquid surface tension.

We can calculate an estimate for \(f_2\) by using the critical capillary number provided in Figure 2 when the contact angle is 90°, by first calculating the critical water velocity using the surface tension
and viscosity for pure water at 20°C, as previously defined. Then by equating Eqs. (5) and (8), we can solve for the drag force correction factor and plot the result. This data is shown in Figure 19.

![Figure 19. Compassion of the experimental drag force correction factor and the value calculated from the literature [13].](image)

### 4 Conclusions

In this paper, the drag force on a 3.9 and 4.6 mm spherical particle attached to an air-water interface under the influence of a stagnation flow field was measured using a specially designed vertical laminar flow column and a modified sphere tensiometry apparatus. The drag force was directly measured after accounting for buoyancy. Drag force correction values over a range of contact angles for the radial component of the modified Stokes drag force equation were determined. The values were calculated from the experimental measurements and compare well to the one other numerical study. The results are also within the same order of magnitude as shown in the limits for the situation when a particle is approaching a bubble and is just touching the surface.
References


Chapter 7

Conclusions and recommendations
1 Contributions to knowledge

In this thesis, I have investigated the detachment of particles by shear flow from a bubble surface. Specifically, I have developed an energy balance model to describe the effect of contact angle hysteresis and bubble deformation during particle detachment, and also directly measured the drag force exerted by laminar water flow, in the normal and tangential directions to an air-water interface, on a stationary floating particle. The achievements made in this thesis are summarised in this section.

In Chapter 3, a novel experimental setup was designed and built for investigating shear flow on the detachment of sub-millimetre spherical particles from a bubble surface. The particle-bubble detachment was captured using high-speed video microscopy and quantified in terms of the particle hydrophobicity (contact angle) and the local water velocity. It was found that a quantifiable hydrodynamic force was able to detach the particles from the bubble surface. The results indicate that there is an increasing trend for the detachment of the particles exhibiting a higher level of hydrophobicity, as indicated by the contact angle, requiring an increased velocity for detachment to occur. It was proven that shear flow can result in the detachment of particles, where there is no oscillation of the bubble surface or particle and where no centrifugal force is exerted.

Chapter 4 was a combination of careful experimental work and corresponding modelling, shedding light on the mechanism of bubble-particle detachment. By choosing a still small but large enough particle size, it was demonstrated exactly how particle detachment occurs from a bubble surface and how this can in fact be fully described by an energy balance model. The research data showed that the contact angle does not remain constant throughout the particle attachment and detachment process. The findings also clearly shows that the bubble profile does not remain spherical as a particle undergoes detachment and provides direct evidence of the pinning and de-pinning of the three-phase contact line. The outcomes of Chapter 4 provide a useful framework for analysing the bubble deformation, interaction force, and change in contact angle during the attachment and detachment using an atomic force microscopy type technique.

In Chapter 5, I directly measured the drag force on different sized spherical particles floating at an air-water interface. A clear correlation was shown between the drag force correction factor for fluid flow in the tangential direction, and the three-phase contact angle. The drag force exerted on the stationary floating particle was directly measured using a carefully calibrated spring wire and a laser triangulation system. The water velocity profile was obtained by using high-speed video microscopy to capture the progress of tracer particles in the channel. The velocities obtained using the high-speed camera were shown to agree with the CFD modelling based on experimental measurements.
In Chapter 6, I directly measured the drag force caused by a stagnation flow field on a floating spherical particle. The drag force was directly measured using a specially designed vertical laminar flow column and a modified sphere tensiometry apparatus. The values were calculated from the experimental measurements and compare well to the one other numerical study. The results are also within the same order of magnitude as shown within the limits for the situation where a particle is approaching a bubble and is just touching the surface.

2 Recommendations for future work

This study represents an ongoing research effort into quantifying the particle detachment process in flotation and the dynamic nature of the forces involved. Despite the substantial research findings in this area, there remains more challenges to fully quantify the detachment of real particles in mineral flotation and to develop models to accurately predict the probability of detachment. For future research, the following recommendations are proposed:

- Results from recorded videos in Chapter 4 studying the bubble deformation during detachment, show that a small air bubble remains on the particle surface after detachment. This indicates that the detachment mechanism involves bridging of the air-water interface and does not completely remove air from the particle surface. This observation is similar to those made by Dippenaar [1] and again more recently by Morris and Cilliers [2], who looked at the effect of particles on bubble coalescence, finding that bridging of the interfaces occurred at a short distance away from a particle.

- During the development of the experimental procedures studying the effect of stagnation flow, a problem of micro-bubbles being distributed throughout the system occurred. These bubbles were observed to stick to the hydrophobic particle surface. It is well known in the literature [3,4] that accumulation of dissolved gases occur on hydrophobic surfaces. Investigation into how the presence of a possible air layer affects the boundary condition for the tangential velocity on the particle surface is thus recommended.

- The effect of particle shape on drag force will be necessary for analysing real flotation systems. Research conducted by Koh et al. [5] found that ground (roughly shaped) ballotini generally has higher flotation rates then spherical (smooth surface) ballotini. In this respect, it will be important to investigate how the drag force changes in response to particle shape.

- It has been demonstrated in Chapter 3 that particle detachment will occur due to shear flow over a bubble surface. It will be important to use the results of Chapters 5 and 6 to develop a model, based on the air hold-up, to predict the probability of particle detachment occurring in coarse particle flotation technologies that utilise fluidisation principles.
References


Appendix A.

Supporting information for

Chapter 4

An analysis of bubble deformation by a sphere relevant to
the measurements of bubble-particle contact interaction and
detachment forces
1 Minimisation of free energy of a pendant bubble interface deformed by a sphere on the top

Here we derive the key equations obtained by minimizing the free energy of a pendant bubble attached to a flat surface and deformed by a sphere on the top. The minimisation of the free energy for this system composes a variation problem with variable end points, viz., the three-phase gas-liquid-solid contact (tpc) points on the particle surface and flat surface can move to minimise the free energy. The tpc on the particle surface, in the system shown in Figure 2, requires special attention because the change in its position is also coupled with the change in the position of the centre of mass of the particle by \( h_c = h_{tpc} + r_p \tan \alpha = h_{tpc} + R_p \cos \alpha \), where \( h_c \) is the height of the centre of the particle mass. Different methods are available to analyse this class of problems. We extend the method used previously for a sphere attached to a planar water surface by Nguyen and Schulze [1].

\[ \tan \cos c \ tpc \ tpc \ tpc \ p h h r h R \alpha \alpha \alpha = \alpha \alpha \alpha + \alpha \alpha \alpha = \alpha \alpha \alpha + \alpha \alpha \alpha, \]

\( c \) in the coordinate system.

Figure 1. Schematic of a bubble attached to both a spherical particle and a solid planner surface. \( r_c \) is the contact radius on the particle surface, \( r_o \) is the bubble contact radius, \( h_{tpc} \) is the height of the meniscus connected to the three phase contact line, \( R_p \) is the particle radius, and \( h \) and \( r \) describe the coordinate system.

Figure 1 shows the geometry of a pendant bubble attached to a planar solid surface which is deformed by a spherical particle on the top in the cylindrical coordinate system \((r, z, \phi)\). The geometry is rotationally symmetrical along the z-axis which is the direction of gravity. As shown in Figure 1, \( h \) describes the height of the interface. The potential energy, \( E_{volume} \), is proportional to the change in the volumes of the three phases involved and their relative positions. The interfacial energy, \( E_{surface} \), is determined by the interface areas and the specific interfacial energies. The potential and
interfacial energies are described by equations (2) and (3) shown below. When minimising the system energies we follow the method utilised by Heady & Cahn [2] through the use of a Lagrange multiplier, λ, with the unit of pressure. It is used to meet the constraint of maintaining the constant bubble volume, $V_b$. The bubble and particle size can be considered to be sufficiently large enough to allow us to neglect the line tension effect. The resulting functional to be minimised can be written as:

$$E = E_{\text{volume}} + E_{\text{surface}} - \lambda V_b$$

(1)

Using the nomenclature defined in Figure 1 we describe the system energies as follows:

$$E_{\text{volume}} = mgh_i + \int_{V_i} P_s dv - \int_{V_j} P_l dv$$

$$= mgh_i \left(1 - \frac{\rho_a}{\rho}\right) - \pi \left(\delta - \rho_a\right) g \left[ h_w \int_0 r^2dh + R_p \frac{3h_c}{3} \left(\frac{2 + 3\cos \alpha - \cos^3 \alpha}{3} + R_p \frac{4}{4} \sin^4 \alpha\right)\right]$$

(2)

$$E_{\text{surface}} = \pi \int h_w \left[2\sigma r \sqrt{1 + r^2} dh + 2\sigma_{tg} \frac{R_p^2}{3} (1 - \cos \alpha) + 2\sigma_{sl} \frac{R_p^2}{3} (1 + \cos \alpha) + \left(A_t - r_0^2\right) \gamma_{sl} + r_0^2 \gamma_{tg}\right]$$

(3)

The bubble volume is described by:

$$V_b = \pi \int_0 h_w r^2dh - \pi R_p^3 \frac{2 - 3\cos \alpha + \cos^3 \alpha}{3}$$

(4)

where $m$ is the mass of particle, $g$ is the acceleration due to gravity, $r' = \partial r / \partial h$, $P_s$ and $P_l$ are the pressures of the air and liquid phases with the density $\rho_a$ and $\delta$, respectively, $\rho$ is the particle density, $\sigma_{tg}$ and $\gamma_{tg}$ are the specific surface energies of the particle-air and flat solid-air interfaces, $\sigma_{sl}$ and $\gamma_{sl}$ are the specific surface energies of the particle-water and flat solid-water interfaces, $A_t$ is the total area of the flat solid-water interface, and $r_0$ is the radius of the flat solid-air contact area.

Inserting equations (2) - (4) into equation (1) gives the following equation for total free energy of the bubble-particle system:

$$E(\alpha, h_{tpc}, r_0) = \pi \int_0 h_w \Phi(h,r,r')dh + \pi \Omega(\alpha, h_{tpc}, r_0) + \text{const}$$

(5)

where the two functional energies are described as follows:
\[ \Phi(h, r, r') = 2\sigma r \sqrt{1 + r'^2} - (\delta - \rho_a) g h r^2 - \lambda r^2 \]  

(6)

\[ \Omega(\alpha, h_{pc}, r_0) = \lambda R_p^3 \left( -3 \cos \alpha + \cos^3 \alpha \right) + \frac{mg}{\pi} \left( h_{pc} + R_p \cos \alpha \right) \left( 1 - \frac{\rho_a}{\rho} \right) \]

\[ - (\delta - \rho_a) g \left\{ R_p^3 h_{pc} + R_p^4 (\cos \alpha - 3) (1 + \cos \alpha)^3 / 12 \right\} \]

\[ + 2 \sigma_s R_p^2 (1 - \cos \alpha) + 2 \sigma_s R_p^2 (1 + \cos \alpha) + r_0^2 \left( \gamma_{is} - \gamma_{sl} \right) \]

(7)

The first variation of equation (5) gives:

\[ \delta(E) = \pi \int \left[ \frac{\partial \Phi}{\partial r} - \frac{d}{dh} \left( \frac{\partial \Phi}{\partial r'} \right) \right] dh \delta r + \pi \left[ \frac{\partial \Omega}{\partial h_{pc}} + \left( \Phi - r' \frac{\partial \Phi}{\partial r'} \right) \right] h_{pc} \delta h_{pc} \]

\[ + \pi \left[ \frac{\partial \Omega}{\partial \alpha} + \left( \frac{\partial \Phi}{\partial r'} \right) h_{pc} \right] \frac{\partial r_{pc}}{\partial \alpha} \delta \alpha + \pi \left[ \frac{\partial \Phi}{\partial r'} + \frac{\partial \Omega}{\partial r'} \right] h_{pc} \frac{\partial r}{\partial \alpha} \delta r_0 \]

(8)

The conditions required for an extremum of the functional \( E \) is that \( \delta(E) = 0 \) for all variations of the limit values. Application of these conditions yields the following set of equations:

\[ \frac{\partial \Phi}{\partial r} - \frac{d}{dh} \left( \frac{\partial \Phi}{\partial r'} \right) = 0 \]  

(9)

\[ \frac{\partial \Omega}{\partial h_{pc}} + \left( \Phi - r' \frac{\partial \Phi}{\partial r'} \right)_{h_{pc}} = 0 \]  

(10)

\[ \frac{\partial \Omega}{\partial \alpha} + \left( \frac{\partial \Phi}{\partial r'} \right)_{h_{pc}} \frac{\partial r_{pc}}{\partial \alpha} = 0 \]

(11)

\[ \left[ \frac{\partial \Phi}{\partial r'} \right]_{h_{pc}} + \frac{\partial \Omega}{\partial r_0} = 0 \]

(12)

Equation (9) is the well-known Euler-Lagrange equation. Substituting the functional \( \Phi(h, r, r') \) into the Euler-Lagrange equation yields the Young-Laplace equation which contains the Lagrange multiplier \( \lambda \) as follows:

\[ \sigma \left\{ \frac{d^2 h / dr^2}{[1 + (dh / dr)^2]^{3/2}} + \frac{dh / dr}{r[1 + (dh / dr)^2]^{3/2}} \right\} = \Delta \rho g h + \lambda \]

(13)

Substituting \( \Omega(\alpha, h_{pc}, r_0) \) and \( \Phi(h, r, r') \) into equation (10) yields the following force balance on the particle:
where $\beta$ is the angle of meniscus inclination to the horizontal line at the tpc location. The left hand side of equation (14) describes the particle weight and the vertical component of the capillary force on the wetting perimeter. These two forces act on the particle in the downward direction as per Figure 1. The three terms on the right hand side of equation (14) describe the upward forces on the particle, i.e., the pressure force (buoyancy) on the gas-solid contact area (projected onto the direction perpendicular to the direction of gravity), the liquid buoyancy on the particle volume immersed in the liquid phase, and the gas (air) buoyancy on the particle volume immersed in the gas phase.

Equation (11), after substituting the required functionals gives the Young equation at the tpc location on the particle surface, which is described by:

$$\sigma \cos \theta + \sigma_{sg} - \sigma_{sl} = 0$$  \hspace{1cm} (15)

where the contact angle on the particle surface satisfies the condition: $\theta = \alpha + \beta$.

Finally, substituting the functionals into equation (12) yields the Young equation for the tpc point on a flat solid surface, which is described as follows:

$$\sigma \cos \theta_0 + \gamma_{sg} - \gamma_{sl} = 0$$  \hspace{1cm} (16)

where $\theta_0$ is the contact angle between the bubble and the flat solid surface.
2 Additional experimental results

(a) Contact angle and forces

(b) Bubble volume and Lambda

Figure 2. Experimental run 1 results for measured force, calculated force and particle contact angle.

(a) Contact angle and forces

(b) Bubble volume and Lambda

Figure 3. Experimental run 2 results for measured force, calculated force and particle contact angle.
Figure 4. Experimental run 3 results for measured force, calculated force and particle contact angle.

Figure 5. Experimental run 4 results for measured force, calculated force and particle contact angle.
Figure 6. Experimental run 5 results for measured force, calculated force and particle contact angle.

Figure 7. Experimental run 6 results for measured force, calculated force and particle contact angle.
Figure 8. Experimental run 7 results for measured force, calculated force and particle contact angle.

Figure 9. Experimental run 8 results for measured force, calculated force and particle contact angle.
Figure 10. Experimental run 9 results for measured force, calculated force and particle contact angle.

Figure 11. Experimental run 10 results for measured force, calculated force and particle contact angle.
Figure 12. Experimental run 11 results for measured force, calculated force and particle contact angle.

Figure 13. Experimental run 12 results for measured force, calculated force and particle contact angle.
References
