Gas Bubble Generation at a Micro-Scale Orifice

By

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Abstract

Around the world, people are faced with the challenge of dealing with cancer every day. The number of new cases of cancer rises every year. In North America alone there were 1.6 million new cases reported in 2002. Many of these cases are treatable, however, there are many types of cancers which are classified as drug resistant tumours. These entities do not respond to chemotherapy drugs as their cell membranes have an innate ability to exclude the drugs. Currently there are no viable treatments for drug resistant tumours which can not be removed surgically. Although this is a dire situation, there has been significant research performed over the last 10 years on the topic of altering cell membrane permeability which could lead to a treatment. By increasing the permeability of the tumour cells using mechanical forces induced by oscillating gas microbubbles excited by ultrasound, it is believed that tumour treatment efficacy can be increased greatly. Artenga Inc. of Ottawa, Ontario in Canada is developing a device which can generate and deliver gas microbubbles and chemotherapy drugs into a tumour in order to treat drug resistant tumours. Their TBI (Transient Bubble Infusion) device will consist of a bubble generator and drug dispensing system. The work in this thesis is a theoretical and experimental investigation into bubble generation at a micro-scale orifice for use in the Artenga TBI. Three experimental scenarios are investigated: Constant gas flow bubble generation in a static liquid, constant gas pressure bubble generation in a static liquid and constant gas pressure bubble generation in a liquid cross flow. The first scenario allows investigation and confirmation of the effects of the previous generated bubble on the next bubble, called the bubble wake effect. The second and third scenarios demonstrate that constant pressure bubble generation at an orifice would not meet the requirements of the TBI device however the technique is useful for researchers working with low concentrations of bubbles. In addition to the experimental work, a theoretical force balance model is developed to predict the size of a generated bubble at a micro-scale orifice under constant applied gas pressure in a static liquid. By taking the compressibility of the gas, dynamic surface tension of the liquid, bubble-orifice contact angle and the bubble wake effect into account, the force balance model successfully predicts the generated bubble size.

To my family and friends who continue to encourage me every day

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Nomenclature

Acronyms:

UCA	Ultrasound contrast agent
RS	Reparable sonoporation
LS	Lethal sonoporation
MI	Mechanical index
TBI	Transient bubble infusion device
MEMS	Microelectromechanical system
GIMP	Graphical image manipulation program

Symbols:

A_b	Bubble surface area
A_o	Orifice cross sectional area
C_d	Drag coefficient
D_b	Bubble diameter
D_{bo}	Bubble diameter before expansion
D_{eff}	Effective orifice diameter
D_{mod}	Drag coefficient modifier
D_N	Nozzle diameter
D_o	Orifice inner diameter
f	Bubble generation frequency

F_b	Force due to buoyancy
F_D	Force due to drag
F_{Dnet}	Net drag force acting on a bubble
F_I	Force on a bubble due to inertia
F_L	Lift force
F_m	Force due to change in momentum
F_σ	Force due to surface tension
g	Acceleration due to gravity
h	Submerged depth
k	Wake velocity constant
m	Mass
M	Equivalent mass of a bubble if it were filled with water
m_b	Mass of a bubble
m_v	Virtual mass of a bubble
$\dot{m}_g$	Mass flow rate of gas
$\dot{m}_o$	Mass flow rate of gas through an orifice
p	Gas pressure
P_{atm}	Atmospheric pressure
p_b	Pressure inside a bubble
p_c	Capillary pressure
P_h	Hydrostatic pressure
P_o	Reservoir pressure
P_{oa}	Absolute reservoir pressure
P_r	Pressure ratio between bubble interior and applied gas pressure
P_σ	Pressure due to surface tension

Q	Gas volume flow rate
Q_{eq}	Equivalent gas flow rate
R	Gas constant
Re	Reynolds number
Re_b	Bubble Reynolds number
t	time
t_b	Bubble formation time
T	Temperature
v_b	Velocity of a bubble rising through a liquid medium
V_b	Bubble volume
v_{CF}	Liquid cross flow velocity
v_o	Gas velocity at pipette exit
v_w	Wake velocity
w/v	Weight per volume (g/ml)
W	Bubble expansion velocity
Z_d	Vertical distance bubble travels before detachment
α	Added mass coefficient
γ	Ratio of specific heats
γ_b	Acceleration of a bubble during growth
θ	Contact angle
μ_w	Water viscosity
ρ_{atm}	Gas density at atmospheric pressure
ρ_g	Gas density
ρ_{gb}	Gas density inside a bubble

ρ_w	Density of water
σ	Surface tension

Chapter 1: Introduction

Around the world cancer is a great concern. Each year the number of new cancer cases grows and the number of lives claimed by cancer grows with it. Worldwide reports suggest that there were 10.9 million new cases of cancer and 6.7 million cancer deaths in 2002. There were 1.6 million new cancer cases reported in North America alone [1].

Many treatment options exist for the various cancers, however these lack efficient drug and gene delivery. Currently, chemotherapy drugs are delivered throughout the body at as high a dose as the patient can withstand. This aggressive therapy improves survival but increases debilitating side effects such as severe anaemia and toxicity. In addition to degrading quality of life, side effects can limit a patient's capacity to withstand further treatment and therefore impact survival.

In addition to the inefficient, uncomfortable treatments, many cancerous tumours are drug resistant. The cell membrane of a drug resistant tumour cell can exclude chemotherapy drugs and therefore prevent destruction of the cell. If a tumour is resistant to treatment and it can not be removed surgically, the results are often fatal. Table 1-1 summarizes some of the major drug resistant cancers, and the number of lives that are claimed by the disease each year in Canada and the United States combined.

Table 1-1. Common drug resistant cancers and the numbers of lives claimed each year in Canada and the United States

Cancer	Drug resistance	Ultrasound guided intervention	Deaths per year in the U.S. and Canada
Pancreas	Inherent	Yes, frequent	34,000
Ovarian	85%	Yes	18,000
Liver	High	Yes, frequent	14,000
Colon	Inherent	Yes	56,000
Breast	60%	Near lungs and ribs	48,000
Total			170,000

Currently there are no viable options available for treating drug resistant tumours which can not be removed surgically. Many researchers are attempting to develop a treatment for drug resistant tumours by attempting to alter cell membrane permeability.

1.1 Manipulation of Cell Membrane Permeability

The manipulation of cell membrane permeability is the focus of intense research because temporarily increasing the permeability of a biological cell could improve drug uptake at the cellular level. This could lead to localized drug delivery, gene therapy and the treatment of currently untreatable drug resistant tumours [1]. The manipulation techniques currently being researched have their origins in the medical imaging field. Ultrasound induced necrosis in sensitive tissue such as the mammalian brain due to high energy ultrasound demonstrated the possibility of using ultrasound energy to manipulate

cells [2]. Initially it appeared the power levels required were excessive, however further studies revealed that at lower energy levels such as those employed in medical imaging ultrasound, cell membrane permeability increased [5]. It was soon observed that the presence of microbubble-based ultrasound contrast agents (UCAs) significantly reduced the power threshold [3]. The phenomenon was dubbed sonoporation [4] or sonophoresis.

Sonoporation can be classified as either reparable sonoporation (RS) or lethal sonoporation (LS) [4]. Reparable sonoporation is defined to be the creation of transient pores in the cell membrane. The pores are considered transient because the cell can repair the damage and return the cell membrane to its original less permeable state. Lethal sonoporation, as the name suggests, causes pores beyond the repair capability of the cell and results in cell death. Closer inspection of the phenomenon in research has revealed several mechanisms that are believed to be responsible for sonoporation. These include gas microjets caused by collapsing microbubbles, shock waves created by oscillating and collapsing microbubbles and shear due to microflows around oscillating microbubbles.

1.1.1 Demonstration of Sonoporation in Research

Lethal sonoporation has been observed in some medical ultrasound imaging scenarios and has raised controversy over the use of UCAs. Wible et al. studied the bioeffects of insonifying the kidneys of a rat test subject [10]. The rat kidneys were insonified for 5 minutes using clinical ultrasound and it was found that up to 40% surface area damage due to renal hemorrhaging occurred at a high mechanical index (MI) of 1.6. Damage was reduced as the MI was reduced and almost no damage occurred below a MI of 0.8. Using an in-situ canine kidney model, Tran et al. demonstrated that, in the presence of

microbubbles the threshold for complete cell lysis was reduced by 30% in some cases [3]. Chen et al. also demonstrated hemolysis by exposure to ultrasound and microbubbles. They further correlated the effects of the ultrasound pulse length and it was shown that longer pulses and higher peak negative pressure cause more hemolysis [11].

Wamel et al. explicitly studied the effect of diagnostic ultrasound on cellular uptake and cell viability. They showed that ultrasound alone can alter cell uptake. The study employed large dye molecules (Texas Red Dextran) which the cell membrane was normally impermeable to. Following the application of ultrasound the number of cells which took up the dye was counted. For a range of ultrasound pressures and duty cycles, they observed trends which suggested high uptake was associated with high cell lysis or permanent damage. The addition of microbubbles increased cell lysis and in this study did not appear to increase uptake. Therefore the conclusion was that increased cell uptake could be achieved with ultrasound alone given that the parameters were manipulated carefully [12]. The fact that high cell lysis occurred suggests that this approach would be better suited for drug resistant tumour treatment as local cell viability is not a concern.

Frenkel et al. also noted that ultrasound on its own enhances cell uptake. More specifically, they noted that the transfer of DNA across cell membranes was increased and that this enhancement was further increased with the addition of microbubbles. They studied the effects of loading albumin shelled microbubbles with DNA as the microbubbles were created. The resulting microbubbles were then sonicated in a test tube with human embryonic kidney cells. It was reported that significant gene expression and

high cell viability was observed in these cells [5]. This increase in transfection shows promise for the application of microbubbles and ultrasound in the gene therapy field.

The most direct measurement of changes in cell membrane permeability due to the application of ultrasound and the presence of microbubbles was carried out by Deng et al [10]. In this study, a *Xenopus* Oocyte (a female gametocyte which an egg develops from) was harvested from a toad. The 0.8 mm diameter cell was then treated and placed in a small tank of specialized solution directly above an ultrasound transducer. Using a microelectrode voltage clamp, the cell's membrane potential was clamped at -50 mV. A schematic of the experimental setup is shown in Figure 1-1. By measuring the transmembrane current, the flow of ions through the membrane was directly measured. A baseline was established and compared to various ultrasound and microbubble combinations [8].

Figure 1-1 Schematic of experimental setup employed by Deng et al.

Deng's study determined that the application of ultrasound without the presence of UCAs did not affect the transmembrane current. However, once UCAs were added to the solution and ultrasound was applied, the transmembrane current increased. This suggests that the membrane becomes more permeable to ion flow into and out of the cell. Once the application of ultrasound ended, the cell membrane permeability returned to its normal baseline state. The time for reparation was on the order of 5 seconds [10].

The stepwise increase in transmembrane current shown in Figure 1-2 demonstrates a definite link between cell permeability and microbubble sonication. The exact mechanism responsible has not been identified, however it has been hypothesized to be a

stress wave – membrane interaction. Several studies have studied the interaction between cell membranes mechanical forces more closely.

Figure 1-2 Membrane permeability under 3 conditions: (a) No ultrasound (US) application; (b) Ultrasound application without microbubbles (MB) present; (c) Ultrasound applied to the cell in the presence of microbubbles [10]

1.1.2 Acoustic Cavitation

In general it is believed that the primary factor that causes sonoporation is microbubble cavitation [1]. In engineering, cavitation is a phenomenon that is most often associated with centrifugal pumps. Small gas pockets or cavities form in the fluid being pumped as it vaporizes due to the low pressure region near the pump inlet. These cavities collapse on the blades of the pump as they flow to higher pressure regions in the pump. The resulting

impingements cause mechanical damage to the blades. Acoustic cavitation is the formation and oscillation of gas pockets due to the addition of acoustic energy such as ultrasound to a fluid. Medical imaging ultrasound frequencies transmit acoustic energy at the resonant frequency of microbubbles of diameter 1 – 10 microns.

Acoustic cavitation has been classified into two categories: non-inertial cavitation and inertial cavitation. Non-inertial cavitation involves continuous oscillation of a gas bubble about its equilibrium radius. Figure 1-3 shows images of a non-intertially cavitating microbubble adjacent to a cell captured using a high speed camera by Wamel et al [4].

Figure 1-3 Non-inertial cavitation. The first frame (top left) highlights the cell and the microbubble. Subsequent frames show the radial oscillation of the microbubble

Inertial cavitation is a growing oscillation of a gas bubble followed by a violent collapse. This violent collapse is accompanied by a radial shock wave and mechanical stresses are induced upon the surrounding environment. If this violent collapse occurs near a wall or beside a cell for example, the microbubble may undergo an asymmetric collapse. Microjets of gas are often produced which impinge on the surface or cell [10,15]. This mechanism is believed to be capable of puncturing a cell membrane. It

should be noted that some researchers have used these terms synonymously with stable cavitation and unstable cavitation however it has been recently revealed that stable inertial cavitation can occur [6].

1.1.3 Physical Mechanisms Responsible for Sonoporation

Acoustic cavitation can cause mechanical damage to a cell through normal stress created by shock waves when a microbubble collapses, shear stress due to microstreaming around a cavitating microbubble or impingement similar to cavitation observed in pumps.

To directly observe the effects of a normal stress wave on cell permeability, Lee et al. designed an experiment that would allow a 300 bar shock wave to strike a cell under observation. By ablating polyimide film in water with a laser, a shock wave can be produced. In the experiments, mouse breast sarcoma cells were placed adjacent to the polyimide film in a liquid medium containing a membrane impermeable fluorescent probe (calcein). A microscope focused on the cells observed the efflux of the probe by noting the intensity of fluorescence. According to the study, the cells' membranes became permeable to the dye following the application of the stress wave. The probe followed a simple time dependent diffusion model which demonstrated that the cell permeability returned to its original state after about 80 seconds. This study successfully shows that the cell membrane permeability is transiently affected by a stress wave while the cell remains viable [13].

The effect of shear stress due to microstreaming around a microbubble has also been studied. Upon exposure to ultrasound, a microbubble may experience radial oscillations. During these oscillations a shearing flow which may compromise the cell membrane

develops adjacent to the bubble. Wu et al. simulated microbubble microstreaming through the use of an oscillating Mason horn. By placing a subject lymphocyte in the focus of the Mason Horn, shear stresses were induced on the membrane of the lymphocyte. Similar to previous studies, the experiment was conducted in a solution containing fluorescent marker molecules. The results demonstrated that at low acoustic pressures (< 4100 Pa) reparable sonoporation can be achieved through shear stress generated by microstreaming. If pressure and shear were increased, lethal sonoporation was observed.

Evidence suggests that mechanical stresses and microjet impingement caused by violent microbubble collapse are also responsible for some changes in membrane permeability. Tang et al. showed that ultrasound induced microbubble cavitation in a liquid medium on the surface of Yorkshire pig skin increases its electrical conductance and hence its permeability. Furthermore, they went on to show that the permeability was directly linked to asymmetric bubble collapse at the skin surface. Their research states that it is possible that shock waves due to inertial cavitation in the bulk medium also cause an increase in cavitation at the surface as they interact with bubbles.

It is likely that a combination of the above mentioned mechanisms is responsible for altering the permeability of a cell membrane in some of the studies that have been performed. The ultrasound frequency, intensity, pulse characteristics, bubble size and cell characteristics appear to determine which mechanism dominates each situation. High energy ultrasound with or without the presence of microbubbles appears to cause high cell lysis and damage. Associated with this high cell lysis is a high drug uptake. It is

possible that microjets, shockwaves and high shear stresses damage many cells beyond repair while some cells with significant damage repair themselves following a period whereby extracellular material is free to enter the cell. Lower energy excitation of the microbubbles and cell membranes causes lower levels of shear stress, fewer shock waves and microjets due to reduced or nonexistent inertial cavitation. It appears that where cell viability is most important the most promising method for increasing cell permeability lies somewhere in between the regime of non-inertial cavitation and inertial cavitation. The transition from one to the other depends on many factors, however it appears from Frenkel et al. that very high cell viability and uptake can be achieved at ultrasound frequencies and pressures where inertial cavitation is expected [5].

1.2 Artenga Inc. and the Need for Microbubble Research

Artenga Inc. began as a one man start-up company in Ottawa, Ontario. Artenga's founder, Mr. James Keenan, believed it was apparent that there was a need to improve tumour treatment. He realized this could be achieved by enhancing medical treatment efficiency by improving cellular drug uptake as many tumours are inherently resistant to drugs employed in traditional chemotherapy. A literature search revealed that researchers had observed that increases in cell membrane permeability could occur when a cell was exposed to diagnostic imaging ultrasound. Furthermore, researchers observed that the phenomenon was enhanced when the cell was exposed to ultrasound in the presence of ultrasound contrast agents. These contrast agents are small gas microbubbles that reflect ultrasound and improve its imaging characteristics. Further literature review revealed, as summarized in the previous chapter, that increases in cell permeability were due to

mechanical interactions between the cells and microbubbles. Mr. Keenan proposed that a medical device could be designed that would use these interactions to controllably alter cell membrane permeability in order to treat drug resistant tumours through direct microbubble and chemotherapy drug injection.

1.2.1 The Transient Bubble Infusion Device (TBI)

Using knowledge gained from literature, design specifications for a device called the Transient bubble infusion device were set out. The medical requirements for the TBI are [13]:

1. Enhance treatment efficacy by improving drug uptake at a cellular level.
2. Permit a high, localized dose of toxic drugs with minimal negative side effects on the target patient.
3. Permit a more flexible, combination treatment than is possible with acoustically activated drugs formulated in pharmaceutical settings. Physicians will be able to administer a variety of drugs (enhanced by acoustic activation), at short notice, tailored to an individual patient's indication, reaction to therapy, and disease stage.
4. Enhance the therapeutic efficacy of acoustically activated drugs. (Acoustically activated drugs are not active until exposed to ultrasound)
5. Be approved for patient use by the appropriate regulatory agency (Health Canada for example).
6. Enhance the ultrasonic visibility of a needle to permit precise, localized drug delivery, to a depth within a patient, under real-time ultrasound guidance.

In order to accomplish these goals, operational design requirements were also outlined:

1. A physician inserts a needle or deploys a catheter to a particular area of interest such as a solid tumour. A drug or combination of drugs is delivered in the form of, or infused with, transient micro or nano bubbles.
2. Ultrasound activates the bubbles by inducing cavitation.
3. Acoustically activating micro or nano bubbles will:
 - a. activate the pharmacological activity of a therapeutic agent, such as enhancing drug transport through tissues and across cell membranes, and, or
 - b. create a local hyperthermic condition that can enhance the destruction of diseased tissue such as cancerous tissue, and, or
 - c. further enhance the drug uptake of acoustically activated drug systems by increasing the local cavitation
4. The TBI device may be used to generate gas bubbles within a liquid carrier. The device may be used with liquefied gas.
5. The device may be used to generate gas or liquid bubbles directly within a patient without a carrier fluid.
6. The bubble size and concentration within a carrier fluid will be adjustable.

The physical properties of the required bubbles are outlined in Table 1-2 [15].

Table 1-2. Required Microbubble Characteristics

Characteristic	Acceptable Range
Nominal Diameter	0.5 – 10 microns
Homogeneity	80% within 50% of mean diameter
Concentration	Minimum 10^6 bubbles/ml
Bubble Stability	Minimum 10 seconds, minutes preferable

A concept sketch of the TBI is shown in Figure 1-4.

Figure 1-4 TBI device concept sketch showing ultrasound transducer, TBI device, and representative injection target [14]

As the project progressed, several more design requirements became evident. It was discovered that the type of gas that comprised the microbubbles had an effect on their performance. Therefore it was determined that the device should allow for the gas type to be changed if required. Biocompatibility and chemical simplicity of the stabilizing agents

was also considered important. The stabilizing agent was required to be biocompatible and require the fewest number of components possible.

1.2.2 Microbubble Generator

The key component of the TBI device is the microbubble generator. The microbubble generator is required to produce microbubbles that meet the requirements set out in Table 1-2. To design such a device, an understanding of the currently available bubble generation techniques was sought. It was determined that there were many ways to generate bubbles, but only one technique allowed bubbles which met the physical requirements to be generated.

The method that meets the physical requirements is the technique that is used to generate microbubbles for use as ultrasound contrast agents. This technique involves the sonication of a gas-liquid mixture.

To prepare the microbubbles, we begin by preparing several phosphate buffer solutions. These solutions are used as a medium to generate microbubbles in and are also used to wash away particulate matter from the microbubble solution. Next, the surfactant or stabilizing agents must be prepared. This usually involves using a mortar and pestle to grind and mix several components within a buffer solution. Immediately after this mixing, the components are autoclaved for about 10 minutes and the ground components are allowed to cool. Once cool, additional buffer solution and the mixture are added to a vial or beaker which has been purged with the gas that will form the microbubbles. Lastly, the mixture is sonicated or exposed to ultrasound for a period of about 5 minutes. The added energy of the ultrasound causes microbubbles to form in the solution.

Following the generation of the microbubbles, the solution must be cooled to a temperature of about 5 degree Celsius. This cooling takes about 30 minutes and allows time for the microbubble mixture to separate into 3 phases. The first phase is a large bubble phase which contains bubbles that have separated out through buoyancy and are too large to be microbubbles. Another phase that is not useful is the bottom phase or the unused surfactant solution. This is a phase which does not contain bubbles. The useful phase is the middle phase in which the microbubbles are located. Once extracted and washed with a buffer solution, the microbubble product can be stored for later use [15].

Due to the length of time required to generate and post-process the microbubbles used for ultrasound contrast agents, it is evident that this method is not suitable for a transient bubble generating device. The method employed must be quick and require little or no post-processing. It was determined that an investigation into bubble generation at an orifice (or an orifice array) submerged in a bubble carrier liquid was required. Since very small bubbles are required for the application, it is expected that the orifice diameter will be on the order of one micron.

Chapter 2: Literature Review

The subject of bubble generation at an orifice has been covered extensively in literature. Many studies have investigated the effects of generating bubbles in reduced gravity, in static liquids, in shear flows and at various orifice angles with respect to gravity. Although the work is extensive, almost every study has been done with orifice or nozzle diameters which are on the order of 1 mm or greater. Only one theoretical study that considered a micro-scale orifice was identified. This study had limited experimental results with which to compare to theory. Several other experimental investigations have been conducted however some experimental details have been withheld, perhaps due to intellectual property concerns. Lastly, a study concerning a MEMS (microelectromechanical system) device was identified. This study also withheld key information about the test setup.

In the current study, bubble generation at a micro-scale orifice submerged in a static liquid and in a liquid cross flow will be investigated. The literature review will serve to provide insight into the physical phenomena that affect the size of a bubble upon detachment. Particular detail will be paid to the forces governing bubble generation in a static liquid.

2.1 Forces acting on a Bubble during Generation at an Orifice in a Static Liquid

There are many common forces that are included in almost every model concerning bubble generation at a large orifice. The forces can be grouped into two categories: attaching forces and detaching forces. Attaching forces, as the name suggests, act to prevent a bubble from detaching from the orifice. Detaching forces act in the opposite direction to promote bubble detachment.

2.1.1 Attaching Forces

The primary attaching forces acting on a bubble during formation are due to surface tension, drag and inertia.

Surface Tension Force

The surface tension force pins the bubble down around the circumference of the orifice. In order to determine the magnitude of the force, pinning the bubble, the surface tension between the liquid and the orifice is integrated about the orifice perimeter. For a circular orifice, this leads to equation 2-1.

$$F_{\sigma} = \sigma \pi D_o \quad (2-1)$$

Drag Force

The drag force on a bubble is defined using the standard expression for calculating drag on an object moving at a given velocity. The expression involves a non-dimensional drag coefficient multiplied by a dynamic pressure and surface area as shown in Equation (2-2).

$$F_D = \frac{1}{2} \rho_w W^2 A_b C_d \quad (2-2)$$

Upon inspection we can see that the drag force on a bubble during formation is not constant. The drag force is proportional to the velocity at which the surface of the bubble moves with respect to the fluid squared. In addition, as the bubble grows its surface area increases and the rate at which its volume increases is reduced. This is generally modelled by assuming an incompressible gas fills the bubble at a constant flow rate. Snabre and Magnifotcham called the velocity at which the bubble rises the “Expansion velocity”, W . Using an empirical value for the distance that a bubble travels upwards before detachment, Z_d (Equation 2-3) and the average time it take for a bubble to form, t_b (Equation 2-4), they derived the average expansion velocity W (Equation 2-5) [16].

$$Z_d = \frac{3D_b}{4} \quad (2-3)$$

$$t_b = \frac{V_b}{Q} \quad (2-4)$$

$$W = \frac{Z_d}{t_b} = \frac{3D_b Q}{4V_b} \quad (2-5)$$

Another important factor affecting the drag force is the selection of the drag coefficient. There are many variations in literature for the value of the drag coefficient. The value selected by researchers depends on the size of the bubbles being generated as well as the result which yields the best fit to experimental data for a given experiment.

The simplest expression often accepted in literature is the Stokesian drag relation which approximates a bubble as a solid sphere [16]. It is generally accepted that the

relation shown in Equation 2-6 is valid for at low bubble Reynolds numbers where there is no internal gas circulation in the bubble [17, 18, 19]. Experimental investigation of the drag coefficient was performed by Miyahara and Takahashi for varying Reynolds number and it was determined that the Stokesian relation over predicted the drag coefficient [20]. Their results are shown in Figure 2-1 and the relation they determined best represented the bubble drag coefficient at low bubble Reynolds number is shown in Equation 2-7.

Figure 2-1 Results of the experimental investigation of bubble drag coefficients by Miyahara and Takahashi [20]

Table 2-1 summarizes accepted bubble drag coefficient values and the region where they have been applied. Many other more complex relations exist however it is noted by Clift that the theoretical predictions rarely agree with experiment. Therefore the drag coefficient used in modelling bubble generation is often selected to best fit the experimental data.

Table 2-1. Accepted bubble drag coefficients

Drag Relation		Applicable Regime	Researcher
$C_D = \frac{24}{Re}$	(2-6)	$Re < 1$	Stokesian [17,18,19]
$C_D = \frac{16}{Re} + 1$	(2-7)	$Re < 1$	Miyahara/Takahashi [20]
$C_D = \frac{18.5}{Re^{0.6}}$	(2-8)	$1 < Re < 1000$	Zhang [21]
$C_D = 0.44$	(2-9)	$Re > 1000$	Zhang [21]
$C_D = \frac{48}{Re}$	(2-10)	$50 < Re < 1000$	Moore [22]

Inertia Force

The inertia force or added mass force takes the inertia of the liquid that the bubble must accelerate as it grows into account. The bubble is considered to have a virtual mass equal to some fraction of M (equation 2-11) which is the mass the bubble would have if it were comprised of the liquid.[19]

$$M = \frac{\rho_w \pi D_b^3}{6} \quad (2-11)$$

Milne-Thomson [23] first derived an expression to represent the virtual mass force, however the term was applicable only to a bubble which is smaller than the orifice. After noting that this case never occurs in area of interest concerning bubble generation, the expression was adapted by Chuang and Goldschmidt. They argued that for a bubble with a diameter larger than the orifice diameter in an infinite stream the virtual mass would lie somewhere between the two values given by Equations 2-12 and 2-13. [19]

$$m_v = M \left(\frac{\rho_g}{\rho_w} + \frac{1}{2} \right) \quad (2-12)$$

$$m_v = M \left(\frac{\rho_g}{\rho_w} + \frac{11}{16} \right) \quad (2-13)$$

They also stated that for cases where the bubble size is greater than 20 times the orifice diameter, Equation (2-12) was applicable. Applying these concepts, Snabre and Magnifotcham derived the relation for the inertia force shown in Equation (2-14), naming an added mass coefficient α . The acceleration of the bubble is given the variable γ_b , and can be obtained by taking the derivative of the velocity in Equation (2-5)[17].

$$F_I = \left(\frac{\rho_{gb}}{\rho_w} + \alpha \right) \rho_w V_b \gamma_b \quad (2-14)$$

2.1.2 Detaching Forces

The detaching forces which are normally considered in literature are the buoyancy force and the force due to change in gas momentum (sometimes called the momentum flux force). In addition to these two forces, a few researchers have proposed a lift force on the bubble being generated due to the wake of the previous bubble that is rising in the liquid.

Buoyancy Force

The Buoyancy force which acts on a bubble is perhaps the simplest force. It exists due to the difference in buoyancy between the gas forming the bubble and the liquid in which the bubble forms. Assuming the bubble is spherical we can derive Equation 2-15.

$$F_b = (\rho_w - \rho_{gb}) g V_b \quad (2-15)$$

Most researchers assume that the gas density is negligible compared to the liquid density and the buoyancy force is therefore given by Equation 2-16.

$$F_b = \rho_w g V_b \quad (2-16)$$

Momentum Flux Force

The momentum flux force exists because the gas entering the bubble is decelerated from the velocity at the orifice to the gas velocity inside the bubble. It is generally assumed that the gas velocity is zero inside the bubble. Sullivan applied Newton's second law and stated that the force due to the addition of mass to the bubble may be obtained through equation 2-17 [24].

$$F_m = v_o \frac{dm}{dt} \quad (2-17)$$

Most researchers simplify this equation by assuming the gas is incompressible. Snabre and Magnifotcham use this assumption and rearrange the expression to yield Equation 2-18.

$$F_m = \frac{\pi}{4} D_o^2 \rho_g v_o^2 \quad (2-18)$$

Wake Effect

A few researchers have noted the possibility of a detaching force due to the influence of the wake behind a bubble on the next bubble being generated. This scenario is depicted schematically in Figure 2-2. In the figure, bubble 1 generates a wake as it rises. The

upward velocity of the liquid drawn in the wake of bubble 1 creates a lift force on bubble 2.

Figure 2-2 Schematic representation of the bubble wake effect. Bubble 1 produces a wake which creates an upward force on bubble 2 as it is being generated

In general the wake term is not mentioned nor considered in current models. If it is, it is usually neglected and therefore there has not been much work done to model the effect.

Chuang and Goldschmidt made an attempt to model the wake effect using a first order analysis. By adapting Schlichting's work in *Boundary Layer Theory* on the boundary layer flows around a sphere, they decided that the velocity defect behind a bubble could be expected to have the form shown in Equation 2-19.

$$v_w = kv_b \left[\frac{\int_0^{\frac{1}{f}} v_b dt}{D_b} \right]^{-\frac{2}{3}} \quad (2-19)$$

By solving the equation of motion for a bubble rising in a liquid, they could solve for the bubble rising velocity, v_b . They reasoned that the velocity defect had the effect of reducing the drag force and in cases of high frequency bubble generation, reversing the effective drag force so that it acted as a lift force. Using this argument they accounted for the wake effect by modifying the drag force expression from Equation 2-2 to yield Equation 2-20.

$$F_D = \frac{1}{2} \rho_w (W - v_w) |W - v_w| A_b C_d \quad (2-20)$$

By inspection we can see that if the wake velocity is greater than the velocity at which the bubble rising during formation, the sign of the drag force will change effectively changing the attaching drag force into a detaching lift force [19].

A similar analysis was carried out by Zeng et al. which employed potential flow theory to estimate the lift force due to a rising bubble. The geometry for the analysis is depicted in Figure 2-3 and the resulting expression is Equation 2-21.

$$F_L = \frac{75}{16} \rho_l W^2 a^2 \left(\frac{a}{H}\right)^8 \quad (2-21)$$

According to Kim et al., this sparked interest in the study of bubble and drop formation at an orifice subjected to a cross flow in microgravity [26]. Both Kim and Nahra et al. demonstrated that by increasing the cross flow velocity, the generated bubble size is reduced. Increasing the gas flow rate tended to increase the size of the bubble generated. The results from Nahra's work suggested that the effect of cross flow was less pronounced for smaller orifice diameters [27,28]. Work by Doku et al. confirmed this trend.

In Doku's work, one of the only studies of microbubble generation at a micro scale orifice was done. The purely experimental work considered the effect of cross flow velocity, applied gas pressure and viscosity on bubbles generated at orifices with 0.4 μm , 0.5 μm , 1 μm , 2 μm and 5 μm inner diameters. Although some details of their experimental setup and results were withheld, their work demonstrated the possibility of generating microbubbles using an orifice [29].

2.3 Additional Factors Influencing Bubble Generation at an Orifice

There are several factors that affect bubble generation at a static orifice that have not yet been mentioned. These include the geometry of the orifice, the wettability of the orifice material and circulatory flows in the bulk fluid.

2.3.1 Orifice Geometry

In literature it is the general practice that bubbles be generated at an orifice with a circular cross section. No studies were found that considered the effect of varying the cross

sectional shape of the orifice. Many studies however note that slight deviations in the geometry could have an effect on the generated bubble size. The effects are not elaborated upon.

The predominant characteristic that has been studied is the difference between bubble generation at an orifice in a plate and a nozzle. Snabre and Magnifotcham studied the effects of placing a circular plate around a nozzle and comparing the resulting generated bubbles with those generated without the presence of the plate (see Figure 2-4).

Figure 2-4 Bubble generation at a plate and a nozzle

The purpose of the plate was to study the effect of reducing large scale recirculation effects. It was determined that the bubble rise velocity was reduced by 8 % and delayed

the bubble detachment resulting in slightly larger bubbles. The change in bubble characteristics was considered small however not negligible [17].

Nahra argued that bubble generation at a wall or plate can vary significantly if there is a liquid cross flow. His work suggested that the surface tension force was significantly lower in the case of an orifice at a wall. This occurs because the bubble is restricted by the wall as part of the bubble downstream of the orifice can make contact with the wall. The result is that the bubble is not completely anchored by the orifice. He concluded that bubble generation at a nozzle involves the full effect of surface tension as shown in Equation 2-1 if the nozzle walls are considered small compared to the orifice diameter. This requirement may not be met when a micro scale orifice is considered [19]. In later work he modelled the reduced “pinning” force due to surface tension in his theoretical analysis of bubble generation in a cross flow [28].

2.3.2 Wettability and Orifice Contact Angle

The Wettability of the material which makes up an orifice or nozzle has been shown to have a significant effect on the generated bubble size. The wettability of an orifice is characterized by the equilibrium contact angle that exists between the orifice material and the liquid. Figure 2-5 shows this schematically. A low contact angle constitutes a system that is highly wettable system whereas a high contact angle constitutes a non-wettable system. For a solid-water system, a solid with a low contact angle is said to be hydrophilic whereas a solid with a high contact angle is said to be hydrophobic.

Figure 2-5 Equilibrium contact angle between a liquid and a solid surface

Lin et al. studied the effect of equilibrium contact angle on bubble generation at an orifice in a plate. They discovered that the bubble base could expand beyond the orifice and subsequently increase the surface tension force. Their work revealed that for a contact angle of less than 55° , the bubble base remained constrained by the orifice. For greater contact angles, the bubble base expanded beyond the orifice size and the final generated bubble size was not a function of orifice diameter. This conclusion was reached by generating bubbles with two different orifice diameters. It was found that bubble sizes converged for high orifice contact angles. An intermediate zone was also identified. In this zone the bubble base expanded but did not expand to the full diameter of the bubble. A schematic depiction of the results is shown in Figure 2-6.

Figure 2-6 Bubble generation domains for different contact angles. Domain I: Hydrophilic. Domain II: Intermediate contact angle. Domain 3: Hydrophobic [30]

Doku et al. considered the effects of liquid hydrophilicity and hydrophobicity on bubble generation at a micro pipette. In their work it was determined that bubble generation in a hydrophobic liquid resulted in the generation of much smaller bubbles. Bubbles were about 3 times smaller and thus for the same gas flow rate, 3 times as many bubbles were generated. They also found that changing the contact angle of the system could reduce the minimum gas pressure required to generate bubbles [29].

2.3.3 Liquid Bulk Recirculation

Liquid bulk recirculation is the circulation of liquid due to the motion of the rising bubble stream. As the bubbles rise from the orifice, they displace liquid and entrain liquid so that near the bubble stream there is a net upward motion of liquid. By continuity this must be balanced by liquid motion in the opposite direction to replace the liquid which has been

drawn up. This would occur away from the bubble stream and as such would create a circulation in the bulk as shown in Figure 2-7.

Figure 2-7 Liquid bulk recirculation due to liquid entrainment into the rising bubble stream

Most studies have not considered bulk recirculation to be significant because the experimental setups are generally situated in relatively large liquid reservoirs. A large reservoir is believed to have very little recirculation due to bubble generation. Snabre and Magnifotcham investigated bulk recirculation effects in a cube liquid reservoir with 20 cm sides and a 0.6 mm diameter orifice. As shown in Figure 2-4, they placed a plate below a nozzle in order to reduce the effects of large scale recirculation. Their results showed that the bubble rise velocity changed by about 8%, bubble detachment was delayed and as a result, larger bubbles were generated. They did not quantify the change in bubble size as it appeared to be small [17]. Zhang et al. also added a plate that they called a stabilizer to reduce circulatory flows. However in their work they did not mention its significance.

2.4 Conclusions Drawn from Literature Review

It is apparent from literature that most studies concerning bubble generation at an orifice have focused on generation at a relatively large orifice. The theoretical models discussed in these works generally balance the force of buoyancy with the forces of drag and surface tension. Although the focus of the works is primarily on these forces, other forces have been considered and attempts to model them have been undertaken. From the insight gained from the literature review, it is believed that a theoretical representation of the process of bubble generation at a micro scale orifice can be produced.

A few studies were found that considered bubble generation at a micro scale orifice. These studies provide insight into the general mechanisms behind the generation and also suggest that it may be possible to generate microbubbles at an orifice. It is clear from literature that the subject has not been investigated well and there is a knowledge gap that must be filled. Although an experimental study has been done for orifice sizes around 1 micron, no theoretical analysis has been found in literature.

In order to determine whether microbubble generation at an orifice is feasible for the TBI device, a thorough investigation into the subject should be completed. This study could use the following insight gained from the literature review. Chaung and Goldschmidt suggested that bubble wake effects will be more pronounced at a smaller orifice [19]. Various authors have proposed a multitude of drag coefficients which could apply for small bubbles. Lin and Doku have highlighted the importance of contact angle of the orifice-liquid system [29,30]. Snabre and Magnifotcham and others have suggested that bubbles generated at a nozzle will be smaller than those generated at an orifice in a

plate [17]. They also suggested that recirculation effects could affect the generated bubble properties. Considering Figure 2-7 we could predict that these effects will be more significant in a small reservoir. Finally, considering the surface tension force in Equation 2-1, we can see that reducing the orifice size and the surface tension should result in smaller bubbles.

Chapter 3: Problem Statement

Currently there are no microbubble generation techniques available that are suitable for the proposed Artenga TBI device. A generation technique that would allow for a high concentration of homogeneous and stable 1 to 10 micron bubbles is required for the device. It is desirable that the generation take place at an orifice submerged in a liquid to preserve device simplicity. A great deal of research has been done on the subject of bubble generation however the TBI device will likely require generation to take place at a micro scale orifice. This is a subject which has not been well studied or documented.

3.1 Objectives

The objective of this work is to investigate the possibility of generating microbubbles at an orifice and attempt to produce a theoretical model to predict the generated bubble size for a given set of conditions. It must also be determined whether this method would be suitable for the TBI device. In addition to these, this work will contribute to the knowledge base and experimental results available on the topic.

3.2 Approach

Two experimental setups will be built in order to investigate the bubble sizes which can be generated. An additional test setup will be employed to investigate the bubble wake

effect on bubble generation at a small capillary or nozzle. Finally a force balance model based on the findings of the literature review and insight from experiments will be formulated.

The experimental setups will employ drawn glass micropipettes. These will serve as micro-scale orifices with inner diameters of 0.5 μm , 1 μm , 2 μm and 5 μm . The first set of experiments will investigate constant gas pressure bubble generation in a static liquid. Secondly, constant gas pressure bubble generation in various liquid cross flows will be investigated. Both experiments will vary orifice diameter, gas pressure and surface tension. The surface tension will be varied through the use of the surfactant Tween 80.

The third experimental setup will be a constant gas flow rate experiment. Using a syringe pump, the frequency of bubble generation will be varied in order to investigate and evaluate the bubble wake effect.

The force balance model will be developed by considering the forces identified in the literature review. By assuming bubble detachment from the orifice once the detaching forces overcome the attaching forces, an algebraic model will be formed. The model will then be programmed into MATLAB and solved. The output from the solution will be imported into Microsoft Excel, analyzed and compared with experimental data.

Chapter 4: Experimental Setup

The experimental apparatuses are designed to allow an investigation into the effect of several key parameters on the size of generated bubbles. As the variables are changed, the changes in generated bubbles are observed through a microscope. The microscope has a camera connected to one of the microscope's eye pieces. Images and video from the camera are displayed on a video screen and a computer monitor. The computer allows images and video to be captured and saved. Using a calibrated image manipulation system, images of bubbles are analyzed to determine their size.

4.1 Equipment

The micro-scale orifice used to generate bubbles in this work is the μ Tip prepulled borosilicate glass micropipette from World Precision Instruments (WPI). A typical pipette is shown in Figure 4-1. In this work it is desirable to vary the orifice diameter, hence pipettes of 0.5 μm , 1 μm , 2 μm and 5 μm inner diameter were employed. The pipette inner diameters are quoted to be within $\pm 20\%$ of the data sheet value. In this work, the pipette inner diameter is tested by measuring the gas flow rate through the pipette at a given pressure.

Figure 4-1 A typical pipette as received from WPI

In addition to variation in the inner diameter, there is also a difference between the geometry of the outer diameter as shown in Figure 4-2. The figure illustrates the difference between a 0.5 μm pipette tip and a 5 μm pipette tip. From the figure it can be seen that the 0.5 μm pipette is much more slender and more gently tapered.

Figure 4-2 The difference between the tip of a 0.5 μm (a) and 5 μm (b) pipette tip

The microscope employed in this work is the MEIJI ML7000 metallurgical microscope shown in Figure 4-3. It is equipped with a 10X or 2.5X eyepiece along with 10X, 20X, 40X and 50X objective lenses. The constant pressure static and cross flow work employs the 10X objective lens coupled with the 10X eye piece (100X magnification) while the constant flow static bubble generation employs the 2.5X eye piece with the 10X objective lens (25X magnification). These microscope configurations coupled with the data acquisition system provide ample magnification to image generated bubbles. Using these lens combinations, the microscope has a working distance of approximately 10 mm. The experimental apparatus must be designed so that the distance between the pipette tip and housing wall is less than the working distance. The x-y-z adjustability of the platform allows for ease of use and adequate flexibility.

Figure 4-3 The MEIJI ML7000 metallurgical microscope

The pressurized gas source employed in the constant pressure static and cross flow generation experiments is a cylinder of pressurized nitrogen. A TurboTorch P250500 regulator is used to control the output pressure. The pressure can be varied between 20 psi and 600 psi, which is more than sufficient for the scope of this work. The regulator output is connected to the pipettes with 1/16" plastic tubing and Swagelok fittings. Figure 4-4 shows the nitrogen tank and regulator.

Figure 4-4 Pressurized nitrogen tank and TurboTorch regulator

The data acquisition system consists of a Hitachi KP-M2U camera mounted on a microscope eye piece, a Hitachi 12" monitor and a computer equipped with a KWorld Security TV capture card and Intervideo image capture software. The image capture software WinDVR allows real time monitoring of the experiment, image capture and video capture through the capture card. WinProducer allows frame by frame analysis of the video captured at 30 frames per second. Figure 4-5 and Figure 4-6 show the components of the data acquisition system.

Figure 4-5 Hitachi camera mounted on microscope eye piece

Figure 4-6 Intervideo WinProducer (a) and WinDVR (b) allow capture and analysis of experimental images and video capture

In order to achieve a constant flow rate for investigation into bubble wake effects, a Braintree BS-8000 syringe pump with a Becton-Dickinson 60 ml syringe was used. The BS-8000 is fully programmable. Coupled with a 60 ml syringe, flow rates varying from 10 $\mu\text{l}/\text{min}$ to 4500 $\mu\text{l}/\text{min}$ can be achieved. By changing the gas flow rate through the micropipette, the frequency of bubble generation can be altered. Low frequency bubble generation will allow the wake effect to be studied. Figure 4-7 shows the syringe pump with 60 ml syringe.

Figure 4-7 Braintree BS-8000 syringe pump and Becton-Dickinson 60 ml syringe

4.1.1 Surfactant: Tween 80

One aspect of bubble generation at a micro-scale orifice that has not been well investigated is the effect of varying liquid surface tension. This can be done through the use of a surfactant in solution. Surfactants are molecules with a polar, hydrophilic head and a non-polar, hydrophobic tail [31]. The hydrophilic head dissolves readily in water

while the hydrophobic tail does not. This property causes the surfactant molecules to gather at gas-liquid interfaces [32]. The surface tension at a gas-liquid interface exists because the molecules at the surface of a liquid have a higher potential energy than the molecules in the liquid bulk. This is because the molecules at the surface have greater attraction to those in the liquid bulk than they do to the gas phase. Surfactant molecule's reduce the surface tension because both ends of the molecule are in their preferred state when they are at an interface. The surfactant tends to decrease the energy between the two phases and thus a reduction in surface tension is observed [32].

The surfactant Tween 80 was selected for this work as Artenga's research has shown that it has synergistic properties when combined with a water insoluble surfactant Span 60. The combination of these surfactants allows small gas microbubbles to be stabilized. In general, small gas microbubbles 10 μm or less in diameter dissolve quickly even in a saturated solution. The combination of Tween 80 and Span 60 prevents this dissolution therefore stabilizing the microbubbles. Since Artenga's future products are likely to employ Tween 80, this surfactant will be used in this work.

Tween 80 is a non-ionic surfactant which is readily dissolved in water. Even small quantities of Tween 80 can reduce the equilibrium surface tension significantly. Figure 4-8 shows the equilibrium surface tension versus concentration in water measured by Callion using a CAHN Dynamic Contact Angle Analyser [33]. The figure also shows a single data point obtained from Schick [34] which shows good agreement with Callion.

Figure 4-8 Equilibrium surface tension versus log of concentration in w/v % (weight/volume %)

Since surfactant molecules reduce surface tension by migrating to an interface, the surface tension as the interface forms is higher than the equilibrium until surfactant molecules move to the interface. The surface tension of a forming interface is known as the dynamic surface tension. The dynamic surface tension of Tween 80 in water was measured by Fuhrling using a bubble tensiometer. Figure 4-9 is adapted from his results.

Figure 4-9 Dynamic surface tension versus time for Tween 80 [35]

The dynamic surface tension should be considered in bubble generation if the bubbles form quickly compared to the time required to reach equilibrium surface tension. The time that it takes for a bubble to form determines how much time surfactant molecules have to migrate to the interface. As can be seen from the figure, if a bubble forms quickly, the surface tension at the interface may be higher than the equilibrium value.

4.2 Apparatus for Bubble Generation at an Orifice Submerged in a Static Liquid

The core of the experimental apparatus, a micropipette mounted in a housing, used to generate bubbles in a static liquid, for both constant pressure and constant flow experiments, is shown in Figure 4-10. In order to allow observation of the bubble generation, a thin housing containing a pipette was constructed for each orifice size

investigated. For each experiment, the pipette holder was mounted on the microscope table, filled with the appropriate solution and connected to the pressurized gas source or syringe pump. For these experiments, the microscope was turned on its side, as shown schematically in Figure 4-11, to allow generated bubbles to rise in the direction of the pipette's longitudinal axis. The difference between the experimental apparatus for the constant pressure and constant flow experiments was the gas source. The constant pressure experiments utilized the pressure cylinder of nitrogen whereas the constant flow experiments utilized the Braintree syringe pump with an air filled syringe.

Figure 4-10 Pipette mounted in housing

Figure 4-11 Schematic of pipette housing mounted on the microscope

4.3 Apparatus for Bubble Generation in a Cross Flow

To generate bubbles in the presence of a liquid cross flow, the experimental apparatus shown in Figure 4-12 was designed and built.

Figure 4-12 Cross flow bubble generation apparatus

The apparatus consists of a liquid reservoir, pump, coarse flow control valve, fine flow control needle valve, coarse particulate filter and a custom milled aluminum flow channel. All components are connected using 0.25" plastic hose. The flow channel is milled into a 4" by 5" by 0.75" block of aluminum. The flow channel itself, which is covered and sealed by a clear plastic window, has a square cross section of 0.125" and is 3" long across middle of the block. At each end of the channel there are hose fittings which allow the connection to the flow loop. In the middle of the flow channel, a 0.125" hole exists perpendicular to the flow channel which allows a pipette to protrude into the flow. A custom aluminum pipette holder connects the pipette to a gas line and allows the pipette position within the milled flow channel to be adjusted. The adjustment is possible because the pipette holder has two O-rings which allow the flow channel to remain sealed as the pipette is adjusted. Figure 4-13 shows the pipette holder. Figure 4-14 and Figure 4-15 show a schematic of the cross flow setup.

Figure 4-14 shows a schematic representation of the three main components unassembled: the milled flow channel, the pipette, and the pipette holder. A schematic of the assembled components is shown in Figure 4-15 where it can be seen that the pipette position can be adjusted through translation of the pipette holder. The dual O-rings incorporated into the design allow the translation while maintaining a seal. The pipette can therefore be positioned in the middle of the cross flow channel.

Figure 4-13 Cross flow pipette holder. Gas line connects to the bottom through NPT fitting and the pipette protrudes from the top. Double O-rings allow the position of the pipette in the flow channel to be adjusted

Figure 4-14 Schematic of unassembled components of the cross flow system

Figure 4-15 A schematic of the assembled cross flow system. As shown in the figure, the pipette holder can translate to adjust the position of the pipette tip. Also shown is an enlarged view of the bubble generation in the flow channel

4.4 Experimental Procedure

The following sections will outline the procedure followed for all experiments performed for bubble generation in a static liquid at constant pressure, constant flow and bubble generation in the presence of a liquid cross flow. Included in the procedure outline are the values at which each experimental variable will be tested.

4.4.1 Constant Pressure Bubble Generation in a Static Liquid

Bubbles were generated at constant pressure at five different pressures, four orifice diameters and in 4 liquid solutions. The gas pressure was varied from 30 psi to 90 psi at 15 psi increments. The orifice diameters employed were 0.5 μm , 1 μm , 2 μm and 5 μm . Bubble generation took place in distilled, deionised water with no surfactant and three surfactant concentrations of 0.001 % w/v, 0.01 % w/v and 1 % w/v.

An experiment began by first choosing the pipette mounted in its housing. After mounting the housing on the microscope table, the housing was filled with the proper solution using a syringe and needle until the liquid level was $\frac{3}{4}$ " above the pipette tip. Once the liquid was injected, the microscope was focused on the pipette tip and the gas pressure adjusted to 30 psi. After a 30 second equilibrium period, a screen capture of the bubble stream was taken using the Intervideo WinDVR software. An example of the screen capture of the bubble stream is shown in Figure 4-16. For the next data point, the pressure was increased by 15 psi and another image was captured. This process was repeated until the pressure reached 90 psi.

Figure 4-16 Constant pressure bubble generation sample data point (0.5 μm pipette, 0.001 % w/v surfactant solution, 30 psi, 100X magnification)

Once a data point was taken for each pressure, the pressure was reduced to 30 psi, the solution was extracted from the housing using the needle and syringe and the next solution was injected. Experiments began with distilled, deionised water and progressed to higher surfactant concentrations to reduced contamination effects. After a pipette was tested with all four solutions and five gas pressures, the entire procedure was repeated with a new pipette size.

Experimental Determination of Gas Flow Rate

In order to verify the theoretical modeling of gas flow rate through the pipette and the actual orifice size, the gas flow rate was measured using the modified bubble generation apparatus shown in Figure 4-17.

Figure 4-17 Schematic of apparatus employed to measure gas flow rate through a pipette

To measure the gas flow rate, the pipette was mounted in a sealed housing which was filled with liquid through a cap in the top of the housing. The housing also had a graduated cylinder attached to the side which was open to the atmosphere. Once the housing was filled with liquid and the cap was replaced, the gas pressure was set to the appropriate value. As the gas flowed through the pipette, liquid was displaced and forced into the graduated cylinder, thus raising the liquid level. By recording the initial and final

liquid levels in the graduated cylinder and the duration of the experiment, the gas flow rate was measured. The experiment was performed in distilled, deionised water for orifice diameters of 1 μm , 2 μm and 5 μm . For the 1 μm orifice the experiment was repeated with surfactant solutions of 0.01% w/v and 1 % w/v to investigate the effect of surface tension on the gas flow rate.

4.4.2 Constant Flow Rate Bubble Generation in a Static Liquid

Bubble generation at constant flow rate was performed to investigate the bubble wake effect. Using a syringe pump, low gas flow rates could be achieved which could not be controllably achieved using the constant pressure regulator system.

After flushing out the pipette housings used for constant pressure experiments with distilled, deionised water, constant flow rate bubble generation was investigated using pipette diameters of 1 μm , 2 μm and 5 μm . Gas flow rates varied from 60 $\mu\text{l}/\text{min}$ up to 650 $\mu\text{l}/\text{min}$ and the experiments were conducted in distilled, deionised water with no surfactants. The setup for the experiments was the same as for the constant pressure experiments except that instead of setting the pressure to a given value, the syringe pump was set to 62 $\mu\text{l}/\text{min}$ and started. Before any bubbles were generated, it took time for pressure to build up in the syringe. Once bubble generation began, a stop watch used to measure the frequency of bubble generation. Once the frequency stabilized over a five minute period, it was assumed that equilibrium had been reached and a data point was taken.

Using the screen capture feature of the WinDVR software, an image of the generated bubble or bubble stream was captured. A sample image is shown in Figure 4-18. For the

1 μm and 2 μm pipette diameters, an excessive amount of time was required to reach the pressure in the syringe required to generate a bubble. To overcome this challenge, at the start of the experiment, the gas flow rate was set to 4000 $\mu\text{l}/\text{min}$ until a bubble was observed. The flow rate was then lowered to the appropriate level and a data point was taken when the frequency stabilized. When a data point was taken, the bubble generation frequency was recorded along with the image of the bubble.

Figure 4-18 Constant flow rate bubble generation sample data point (1 μm pipette, distilled, deionised water, 65 $\mu\text{l}/\text{min}$, 20X magnification)

4.4.3 Bubble Generation in a Liquid Cross Flow

Bubble generation in a liquid cross flow was performed to determine whether it is feasible to generate small bubbles at an orifice at constant gas pressure. Similar to the bubble generation in a static liquid experiments, the liquid cross flow experiments varied the orifice diameter from 0.5 μm to 5 μm , the gas pressure from 30 psi to 90 psi and the

surfactant concentration from 0 % to 1 % w/v. In addition to this, data points were taken at average liquid flow velocities of 10 cm/s, 20 cm/s, 40 cm/s, 80 cm/s and 140 cm/s.

The first step in a liquid cross flow experiment was to insert the proper pipette into the custom pipette holder and position it in the flow channel such that the tip of the pipette was in the centre of the channel. Once this was done, the pipette was connected to the gas source and the gas pressure was set to 30 psi. The gas pressure increased before liquid was in the flow channel to prevent liquid being drawn into the pipette due to capillary flow. After the gas pressure was applied, the liquid reservoir was filled with 550 ml of solution. The coarse flow valve was then shut off and the pump was started. The coarse flow valve was then opened slightly to allow liquid to fill the filter, flow loop and flow channel. The liquid flow rate was limited in order to minimize the impact of the liquid on the fragile pipette tip when it reached the flow channel. If the flow rate was too high, the liquid in the flow channel could fracture the pipette tip upon initial contact.

Once liquid was present in most of the flow loop, the flow rate was increased by opening the coarse flow valve. Bubbles that were trapped in the lines were then purged by moving the flow lines by hand to encourage the bubbles to flow through the loop and out into the open reservoir. Care was taken to lower the flow rate when large bubbles passed through the flow channel to avoid damaging the pipette. After most of the air was removed from the system, the coarse flow valve was opened completely and the liquid flow rate was reduced using the fine control needle valve. To adjust the flow rate to the level corresponding to the desired liquid velocity, the needle valve was adjusted and the

flow rate was measured using a 100 ml graduated cylinder and a stop watch. This was repeated until the flow rate was set to the proper level.

After the experiment was set up as described above, the data points could be taken. With the pressure set to 30 psi, the proper pipette and solution in the apparatus, the flow rate was set to correspond to a cross flow velocity of 10 cm/s. Once an image such as the one shown in Figure 4-19 was captured, the flow rate was increased to correspond to 20 cm/s cross flow velocity and another data point was taken. This was repeated until the maximum flow rate was reached. The pressure was then increased to 45 psi and the flow rate reduced to correspond to a cross flow of 10 cm /s. When a data point was captured for every pressure and cross flow velocity desired for a given liquid solution and pipette diameter, the pump was turned off and the liquid was drained from the system.

Figure 4-19 Cross flow bubble generation sample data point (0.5 μm pipette, distilled, deionised water, 30 psi, 20 cm/s, 100X magnification)

For the next set of data points, a new solution was introduced into the system and data points were taken for the varying pressures and cross flow velocities. When data points had been taken for all four solutions, the pipette was removed and a new pipette of larger diameter was inserted. The experimental procedure was repeated for each of the four pipette diameters. Similar to the static bubble generation experiments, the first solution to be tested was the distilled, deionised water. The next solution always contained a higher surfactant concentration than the previous experiment in order to minimize contamination. Between experiments in which the pipette was changed, the flow loop was purged by passing 2 litres of distilled, deionised water through the system in order to remove surfactant which was trapped in the system.

4.5 Data Processing

After the experiments were completed, the data and images captured had to be organized and processed. The numerical data gathered such as experiment parameters and results parameters such as bubble generation frequency were tabulated in Microsoft Excel. Images were numbered to correspond to the experimental parameters under which they were taken. Most of the post processing work involved measuring the size of the bubbles by determined the physical size of a pixel in the captured image through the use of a calibrated scale. A scale which corresponds to the size of an image on the 12" Hitachi monitor was used to correlate on screen size to actual size. By viewing a metallography sample and determining the size of a given feature in the image, the size of the feature in a screen capture image from WinDVR would allow the determination of the size of one

pixel. By using this value of length for one pixel, the size of a bubble or bubble stream could be determined from image captures.

Using a graphical image manipulation program called GIMP and Microsoft Paint, the images were analyzed. Figure 4-20 shows the original metallographic sample image used to transfer the scale from the 12" Hitachi viewing monitor to the captured image size. Figure 4-21 shows the image used to determine the pixel size for constant flow experiments. A 1 mm diameter glass tube is imaged to determine the size.

Figure 4-20 Original calibration image used to determine the size of 1 pixel for cross flow and constant pressure bubble generation experiments

Figure 4-21 Original calibration image used to determine the size of 1 pixel for constant flow bubble generation experiments

In order to process an image, the image was rotated using GIMP so that the bubble stream was approximately horizontal. If the image was of sufficient quality, the image was then exported to Paint. In Paint, lines were drawn using the line tool in order to approximate the boundary of the bubble in the image. Using the X-Y coordinate system, the size of the bubble could be calculated from the difference in the y coordinate of the lines. Figure 4-22 shows an example of the processing of a bubble image of sufficient quality. If the image was not of sufficient quality, filters were applied to the image using GIMP such that the contrast and brightness allowed the bubble size to be measured. An example of this is shown in Figure 4-23. Once the size was measured for each image, the data was tabulated in Microsoft Excel.

Figure 4-22 Captured image shows bubble size measured in Microsoft Paint

Figure 4-23 Captured image after filters applied with GIMP. Vertical line added with 10 pixel segments

Chapter 5: Experimental Results and Analysis

5.1 Introduction

This chapter presents the results and analysis of the bubble generation experiments for all three sets of experiments. The first results to be presented will be the results of the constant pressure bubble generation in a static liquid. Figures depicting the bubble size versus applied gas pressure will be presented for varying surfactant concentrations and orifice diameters. Secondly the results for bubble generation at a constant gas flow rate in a static liquid will be presented. Figures depicting the relationships between bubble size, gas flow rate and bubble generation frequency will be presented. Lastly the results of constant pressure bubble generation in a liquid cross flow will be presented. These figures will depict bubble size generated for various cross flow velocities, applied gas pressures, orifice diameters and surfactant concentrations. An analysis of the trends observed in the experiments will follow each set of results. Since a large amount of data was collected concerning this topic and therefore representative data at a gas pressure of 60 psi and a cross flow velocity of 60 cm/s will be included in this chapter. Some key figures will be included in addition to these with all other data being included in Appendix A.

5.2 Constant Pressure Bubble Generation in a Static Liquid

For these experiments, the generated bubble size at various applied gas pressures was observed for various surfactant concentrations and orifice diameters.

5.2.1 Effect of Varying Surfactant Concentration for Four Orifice Diameters

Figure 5-1 through Figure 5-4 show the generated bubble diameter versus applied gas pressure for four different surfactant concentrations. Each figure shows the results for a different orifice diameter.

Figure 5-1 Bubble size versus applied gas pressure for four surfactant concentrations for an orifice diameter of $0.5 \mu\text{m}$

Figure 5-2 Bubble size versus applied gas pressure for four surfactant concentrations for an orifice diameter of 1 μm

Figure 5-3 Bubble size versus applied gas pressure for four surfactant concentrations for an orifice diameter of 2 μm

Figure 5-4 Bubble size versus applied gas pressure for four surfactant concentrations for an orifice diameter of 5 μm

Upon inspection we can see that the trend in bubble size is similar for all four orifice diameters. The bubble size increases almost linearly with pressure. The effect of increasing pressure on bubble size appears to decrease slightly as the slope decreases above 60 psi for all cases. From the above figures it can also be seen that the effect of decreasing surface tension by increasing surfactant concentration appears to diminish as the orifice diameter increases. This is evident when we compare the bubble size generated in distilled, deionised water versus the bubble size generated in a 1 % w/v surfactant solution as shown in Table 5-1.

Table 5-1 Effect of surfactant concentration decreases with increasing orifice diameter

Orifice Size (μm)	D_b (μm) (0% w/v)	D_b (μm) (1% w/v)	Change in D_b
0.5	215	120	44.2%
1	325	232	28.6 %
2	332	285	14.2 %
5	422	395	6.4 %

This trend could be explained by considering the force balance between attaching and detaching forces to be only between buoyancy and surface tension (something that is commonly done for large orifice diameters around 1 mm in diameter)[19]. Consider the fact that the attaching surface tension force varies linearly with the orifice diameter while the detaching buoyancy force is proportional to the bubble diameter cubed. If we assume the bubble diameter is roughly a linear function of the orifice diameter then we can see that the surface tension force would be less significant as the orifice diameter increases.

An anomaly is observed for the smaller orifice diameters at low surfactant concentrations. We see that at low pressures the bubble size generated is much larger than what would be expected when we consider the trends of the data. These large bubbles are observed when the pressure is near the minimum gas pressure required to overcome the capillary pressure of the orifice. When the pressure is low, the bubble size can oscillate between large bubbles at low generation frequency and smaller bubble at high generation frequency. If the pressure is increased slightly, the smaller bubble size is

generated consistently and the data agrees with the trends. This phenomenon may be due to small geometric variations in the pipettes which are overcome when the surface tension is lowered or the gas velocity is increased to increase the detaching forces.

5.2.2 Effect of Varying Orifice Diameter for Four Surfactant Concentrations

Figure 5-5 through Figure 5-8 show the generated bubble diameter versus applied gas pressure for various orifice diameters. Each figure represents data taken at a different surfactant concentration.

Figure 5-5 Bubble size versus applied gas pressure for four orifice diameters with a surfactant concentration of 0% w/v

Figure 5-6 Bubble size versus applied gas pressure for four orifice diameters with a surfactant concentration of 0.001% w/v

Figure 5-7 Bubble size versus applied gas pressure for four orifice diameters with a surfactant concentration of 0.01% w/v

Figure 5-8 Bubble size versus applied gas pressure for four orifice diameters with a surfactant concentration of 1% w/v

The above figures show that a similar trend exists for bubble size versus orifice diameter for all four surfactant concentrations considered. At low surfactant concentrations the 1 μm and 2 μm orifices generated similarly sized bubbles. In fact for a 0.001% concentration, in some instances the 1 μm orifice generated larger bubbles than that 2 μm orifice. It is possible that this was caused by a leak in the experimental apparatus. The 1 μm pipette holder had a leak for the first two tests. The leak could have cause the water level to drop thus causing a downward flow of liquid to tend to cause the bubbles to remain attached and become larger before detachment. The larger bubbles could also be caused by a unique feature of the 1 μm orifice geometry which is less influential at lower surface tensions.

By plotting the bubble size versus orifice diameter for several gas pressures for a given surfactant concentration, we can gain further insight into the effect of the orifice diameter. Figure 5-9 and Figure 5-10 show bubble size versus orifice diameter for a surfactant concentration of 0.01% w/v.

Figure 5-9 Bubble size versus orifice diameter at several gas pressures for a surfactant solution of 0.01% w/v

Figure 5-10 Bubble size versus orifice diameter at several gas pressures in distilled, deionised water

The figures above indicate that bubble size does not vary linearly with orifice diameter. In general it is expected that the bubble diameter will vary with the cube root of the orifice diameter if we consider the bubble size to be strictly a function of buoyancy and surface tension. Combining Equations 2-1 and 2-16 we get:

$$D_b = \sqrt[3]{\frac{6D_o\sigma}{g\rho_w}} \quad (5-1)$$

Figure 5-11 shows the expected trend obtained by fitting the value of bubble size for the 5 μm orifice from experimental data.

Figure 5-11 Expected and actual trends for bubble size at a given orifice diameter

From the figure above, we see that the trend observed in the experiment is similar to this expected trend however the change in bubble diameter is more pronounced than expected. Analysis of the images captured during the experiments has revealed that the contact angle between the bubble and the orifice may be the cause.

5.2.3 Variation of Contact Angle with Orifice Diameter

During this experimental work, it was observed that the contact angle that the bubble makes with the orifice during formation varied with the orifice diameter. The contact angle also varied with pressure and surfactant concentration although these were less significant. Figure 5-12 and Figure 5-13 show the contact angles measured using the image analysis software GIMP and Paint for 0% w/v and 1% w/v surfactant solutions respectively. GIMP includes a tool which allows the angle between two lines to be

measured. The lines are drawn with Paint to correspond to the angle that the bubble makes with the orifice during generation.

Figure 5-12 Contact angle measurements for bubbles generated in distilled, deionised water

Figure 5-13 Contact angle measurements for bubbles generated in a 1% w/v surfactant solution

As can be seen from the figures above, the contact angle varies significantly between orifice sizes. This is important because the surface tension force that is pinning the bubble acts in the vertical direction. For simplicity, most models assume that the contact angle is zero. This is almost always the case for large orifices however as the figures show, the contact angle for small orifice diameters is often not zero. The surface tension force will therefore be reduced by the cosine of the angle. This concept is depicted schematically in Figure 5-14.

Figure 5-14 Schematic of the reduction of the surface tension force due to the contact angle

The figure above shows the surface tension force, F_σ , broken into vertical and horizontal components, $F_{\sigma y}$ and $F_{\sigma x}$. From the figure we can see that the x-components of the surface tension force will not contribute to pinning the bubble to the orifice and therefore the surface tension force will be reduced by the cosine of the contact angle. Therefore the true surface tension force will be given by:

$$F_\sigma = \pi D_0 \sigma \cos(\theta) \quad (5-2)$$

5.2.4 Variation of Gas Flow Rate with Applied Gas Pressure

In order to model the formation of bubbles at constant pressure in a static liquid, we must have an accurate representation of the forces involved. The frequency of bubble generation and gas momentum both affect the bubble size and therefore the gas flow rate through the orifice must be known. In the next chapter a theoretical calculation of the gas flow rate at different gas pressures and orifice sizes will be performed. To verify this theory, an experiment to measure the gas flow rate was performed. In addition, since the

pipette inner diameter quoted by the manufacturer is $D_o \pm 20\%$, the experiment will also be used to verify the inner diameter of the pipette.

Figure 5-15 through Figure 5-17 show the results of the gas flow rate experiments compared to the theory from the next chapter. The orifice size employed in the theoretical calculation is chosen to best fit the data.

Figure 5-15 Experimental and theoretical results for gas flow rate versus pressure for a 1 μm pipette

Figure 5-16 Experimental and theoretical results for gas flow rate versus pressure for a 2 μm pipette

Figure 5-17 Experimental and theoretical results for gas flow rate versus pressure for a 5 μm pipette

The orifice diameter chosen for the theoretical calculation was chosen as a best fit because the pipette manufacturer quoted the orifice size of each pipette to be $D_0 \pm 20\%$. The experimental data was therefore used to determine the actual orifice diameter. The theoretical results were verified using the Lenox Laser flow calculator [36]. Reasonable agreement was found between the theory and the flow calculator.

The results of the gas flow rate experiments suggest that the actual orifice sizes of the pipettes may be larger than the manufacturer suggests. It must be concluded that the orifice sizes suggested by the theory are correct as only the orifice size and gas pressure can affect the gas flow rate when the flow is choked. Theoretical analysis of the bubble generation will employ the best fit values for the orifice diameters.

Figure 5-18 below shows the effect of varying the surface tension on the gas flow rate. It is evident from the figure that, as we expect, the gas flow rate is not affected by the surface tension.

Figure 5-18 Variation of gas flow rate with surfactant concentration for a 1 μm pipette

5.2.5 Variation of Bubble Generation Frequency with Applied Gas Pressure

It is desirable to know the bubble generation frequency so that the time required for the bubble surface to form may be known. From this data, the dynamic surface tension value can be obtained. From the measured values of gas flow rate and bubble size, using continuity, we can calculate the bubble generation frequency. By dividing the mass flow rate of the gas through the orifice by the mass of one bubble, we get:

$$f = \frac{\dot{m}_g}{m_b} = \frac{\rho_g v_g A_o}{\rho_{atm} V_b}$$

The figures below show the generation frequencies for bubble generation at 1 μm , 2 μm and 5 μm orifices in a 0.01% w/v surfactant solution. The experiment for 0.5 μm was not successful as the orifice was blocked.

Figure 5-19 Bubble generation frequency versus pressure for a 1 µm orifice submerged in a 0.01 % w/v surfactant solution

Figure 5-20 Bubble generation frequency versus pressure for a 2 µm orifice submerged in a 0.01 % w/v surfactant solution

Figure 5-21 Bubble generation frequency versus pressure for a 5 μm orifice submerged in a 0.01 % w/v surfactant solution

5.3 Constant Gas Flow Rate Bubble Generation in a Static Liquid

For these experiments, the generated bubble size at various gas flow rates was observed for various orifice diameters. The purpose of these experiments was to gather data to investigate the bubble wake effect.

5.3.1 Effect of Gas Flow Rate on Generated Bubble Size

Figure 5-22 through Figure 5-24 show the generated bubble diameter versus gas flow rate. Each figure represents data taken at a different orifice diameter.

Figure 5-22 Generated bubble size versus gas flow rate for a $1\ \mu\text{m}$ orifice diameter

Figure 5-23 Generated bubble size versus gas flow rate for a $2\ \mu\text{m}$ orifice diameter

Figure 5-24 Generated bubble size versus gas flow rate for a 5 μm orifice diameter

The figures above show that bubble size decreases as gas flow rate increases until a certain point where the bubble size undergoes a jump in size. During the experiments it was observed that the bubble size increase at this point was abrupt. The bubble size would decrease slowly as flow rate increased and then suddenly the bubble size would begin to oscillate between the large bubble size and small bubble size even as the flow rate remained constant. If the flow rate was increased slightly, the bubble size would remain large and if the flow rate was decreased slightly, the generated size would be small. This jump in size was observed for all orifice diameters tested however for the 1 μm orifice, the jump occurred at flow rates higher than those observed with the other orifices.

The slow decline in bubble size with increasing gas flow rate can be explained by two possible factors. The first is due to the fact that the gas momentum is increasing as the flow rate is increased. Since the change in gas momentum entering the bubble causes a detaching force, this increase could explain the smaller generated bubbles. The second possibility is a lift force generated on each subsequent bubble by the previous bubble's wake. Chuang observed similar results for a 13.6 μm diameter glass orifice [37]. In his work the generated bubble size was observed for varying gas flow rates of up to 340 $\mu\text{l}/\text{min}$. It was noted that bubble size decreased gradually as gas flow rate increased. This trend was attributed to the lift force generated on a bubble due to the previous bubble rising through the liquid medium. By considering Schlichting's work on boundary layer theory, the explanation was justified and the mathematical relationship described in section 2.1.2 was proposed. By considering Figure 4-18 below, we can see that the trend observed by Chuang [37] is similar to the trend observed in this work.

Figure 5-25 Bubble size versus flow rate results from Chuang's work

It should be noted that the capillary pressure for a 13.6 μm orifice is much lower than that of a 1 μm orifice ($p_c(13.6 \mu\text{m})=1.5 \text{ psi}$, $p_c(1 \mu\text{m})= 21 \text{ psi}$). As such, the gas flow rate through the 1 μm orifice during the generation of one bubble is not constant because the pressure at the pipette tip must build up before the gas can flow through the orifice. This fact could cause the gas momentum to be a factor for the smaller orifice sizes in this work whereas it is negligible for the flow rates considered in Chuang's work [19].

Since the bubbles become smaller as the gas flow rate increases, the frequency of bubble generation must increase. This means that the bubbles must be closer together which would cause the wake effect to be more pronounced. Figure 5-26 through Figure 5-28 show the bubble size versus bubble generation frequency for the various orifice diameters and the gas flow rates corresponding to the figures above. From these results, it

is evident that the wake effect phenomenon may play a part in the formation of the gas bubbles.

Figure 5-26 Bubble diameter versus bubble generation frequency for a 1 μm orifice

Figure 5-27 Bubble diameter versus generation frequency for a 2 μm orifice

Figure 5-28 Bubble diameter versus generation frequency for a 5 μm orifice

5.3.2 Bubble Wake Flow Visualization

The flow behind a rising bubble can be visualized by tracing the flow with particles which follow the flow. The Artenga Microbubble Generating Device (MGD) is capable of generating microbubbles with a diameter between 1 μm and 10 μm . The size of these microbubbles dictates that they have very little buoyancy and hence do not rise in the liquid. Any microbubbles below 10 μm in diameter follow the liquid flow with little resistance and act as flow tracers. By injecting the MGD microbubbles into the bulk liquid during a constant gas flow rate bubble generation experiment and performing a frame by frame analysis of the video captured, the flow behind a rising bubble can be visualized.

In this experiment, the MGD was used to generate 3 ml of 1 μm – 5 μm bubbles at a concentration of approximately 10^7 bubbles/ml. The constant gas flow rate bubble generation technique was employed and the microbubbles were injected into 25 ml of 0.01% w/v Tween 80 liquid bulk. Video was recorded for bubble generation frequencies of 0.5 bubbles/s, 1.5 bubbles/s, 4 bubbles/s and a high frequency of about 30 bubbles/s. The figures below show the movement of microbubble flow tracers near a generated bubble for each of the frequencies. The figures show four frames captured $1/30^{\text{th}}$ of a second apart. The path of two circled particles A and B are followed in each image. Particles A and B are at rest in frame 1 except for the high frequency bubble generation figure. A datum line marks the starting point of each particle.

Figure 5-29 Bubble wake flow visualization for a generation frequency of 0.5 bubbles/s

Figure 5-30 Bubble wake flow visualization for a generation frequency of 1.5 bubbles/s

Figure 5-31 Bubble wake flow visualization for a generation frequency of 4 bubbles/s

Figure 5-32 Bubble wake flow visualization for a generation frequency of about 30 bubbles/s

From the figures above we can see that a bubble which has detached from an orifice will induce a flow in the liquid medium around the orifice. If the generation frequency is low enough, the previous bubble will not affect the next bubble because the liquid will come to rest before the next bubble is formed. As the generation frequency increases, the effect on the next bubble increases. The reason for this is two-fold: First, if the generation frequency is high, the distance between the previous bubble and the next bubble is small. Secondly, at high frequencies (greater than 10 bubbles/s) the liquid medium moves

continuously rather than in pulses. When this occurs, the liquid medium is no longer static and hence the lift force due to the liquid will promote bubble detachment. From Figure 5-32, we can see streaks on the image due to particles which are moving quickly because they are entrained in the rising bubble stream. The generation frequency in this image is small compared to the frequencies which occur in the constant pressure bubble generation experiments. Frequencies in excess of 500 bubbles/s are encountered in these experiments; therefore we must conclude that the lift force due to the bubble stream is significant.

5.4 Constant Pressure Bubble Generation in a Liquid Cross Flow

The cross flow experiments yielded a large set of data since cross flow velocity, gas pressure, orifice diameter and surfactant concentration were varied. The following section will summarize the results by presenting the key trends through the use of figures depicting representative data. This data is taken from the experiments involving a liquid cross flow velocity of 60 cm/s and a gas pressure of 60 psi at various orifice diameters and surfactant concentrations. In addition to this, additional figures will be presented to illustrate other key trends. The full set of experimental data can be found in Appendix A.

5.4.1 Effect of Cross Flow Velocity on Bubble Size

To investigate the effect of the liquid cross flow velocity on bubble size, we can plot the bubble size versus cross flow velocity for a constant gas pressure, orifice diameter and surfactant concentration. The results for a gas pressure of 60 psi are representative of the

overall trend. Figure 5-33 through Figure 5-36 show the results for various orifice diameters and surfactant concentrations.

Figure 5-33 Bubble size versus cross flow velocity for a $0.5 \mu\text{m}$ orifice, 60 psi gas pressure and various surfactant concentrations

Figure 5-34 Bubble size versus cross flow velocity for a 1 μm orifice, 60 psi gas pressure and various surfactant concentrations

Figure 5-35 Bubble size versus cross flow velocity for a 2 μm orifice, 60 psi gas pressure and various surfactant concentrations

Figure 5-36 Bubble size versus cross flow velocity for a 5 μm orifice, 60 psi gas pressure and various surfactant concentrations

From the results above we can see that the bubble size is very sensitive to the liquid flow velocity at low cross flow velocities and less sensitive at high cross flow velocities. To understand the reason for this, we must first consider the mechanism that causes the bubble size to decrease. The liquid cross flow promotes bubble detachment through two modes. The first is a drag force on the bubble which causes the bubble to be pulled away from the orifice. This drag force acts to overcome the surface tension force which is pinning the bubble to the orifice. When the flow velocity is low, the drag coefficient is large. At low flow velocities, the bubble, and hence its surface area, is also large. The drag force acting on a bubble is proportional to the surface area of the bubble and the drag coefficient. At low flow velocities, the drag force is large but as the flow velocity

increases and the bubble gets smaller, the drag coefficient and bubble surface area decrease which causes the increase in the drag force detaching the bubble to be small. Figure 2-1 showed the variation of drag coefficient versus Reynolds number and we can see that an increase in flow velocity (Reynolds number) results in a decrease in drag coefficient. The second mechanism that acts to detach the bubble because of the cross flow is a shear force that tends to reduce the surface tension force and close the bubble neck to prevent gas from entering the bubble [27].

From the results presented above, we can also conclude that the effect of the cross flow velocity is more pronounced for bubble generation at a small orifice. This could be due to the fact that the surface tension force is larger for a larger orifice. Since the surface tension force is proportional to the diameter of the orifice, a larger orifice would pin a bubble with a larger force.

Lastly, we conclude that the surfactant concentration is less of a factor at high cross flow velocities than at low velocities. It is observed that the variation in bubble size for different surfactant concentrations is quite large at low flow velocities however as the flow velocity increases, the bubble sizes converge. This can be attributed to the fact that the magnitude of the surface tension force decreases compared to the magnitude of the detaching drag and shear forces due to the cross flow. Therefore the value of surface tension is no longer significant in the force balance and the bubble size is determined by the magnitude of the drag force.

When comparing the experimental results to those found in literature, we can see that similar trends have been observed in literature. Doku observed the generation of bubble in a distilled water cross flow. His work did not divulge all of the details of the experiment however we can see from Figure 5-37 that similar trends were obtained. The results in the figure were taken at an unknown gas pressure.

Figure 5-37 Bubble size versus cross flow velocity presented by Doku [29]

Nahra also investigated bubble generation in a cross flow however his work involved 0.15 cm and 0.076 cm orifices. The investigations considered the effects of low gravity and compared the results in low gravity to those under terrestrial conditions. By observing Nahra's work, we can see that the results obtained for this work at a micro-scale orifice resemble those of Nahra's work in low gravity. Figure 5-38 shows Nahra's results in low gravity and Figure 5-39 shows the results obtained under terrestrial conditions.

Figure 5-38 Bubble size versus cross flow velocity in low gravity as reported by Nahra [28]

Figure 5-39 Bubble size versus cross flow velocity under terrestrial conditions as reported by Nahra [28]

It is possible that the results of the current work resemble those of Nahra's work in low gravity because the buoyancy force for small bubbles is small compared to the drag force due to the cross flow. The buoyancy force is proportional to the bubble diameter cubed while the drag force is proportional to the bubble diameter squared and the surface tension force nearly directly proportional to the bubble diameter. From the above results it appears that for bubbles above 1 mm in diameter, the buoyancy force is dominant however for small bubbles, it is less significant. This could explain why this work resembles the results in low gravity [27,28].

5.4.2 Effect of Applied Gas Pressure on Bubble Size

By plotting bubble size versus applied gas pressure at a constant cross flow velocity of 60 cm/s and various surfactant concentrations for several orifice diameters, we can

investigate the effect that the gas pressure has on the bubble size. Figure 5-40 through Figure 5-43 show the results and the trends which are representative of the experimental data.

Figure 5-40 Bubble size versus applied gas pressure for a 0.5 μm orifice and a cross flow velocity of 60 cm/s

Figure 5-41 Bubble size versus applied gas pressure for a 1 μm orifice and a cross flow velocity of 60 cm/s

Figure 5-42 Bubble size versus applied gas pressure for a 2 μm orifice and a cross flow velocity of 60 cm/s

Figure 5-43 Bubble size versus applied gas pressure for a 5 μm orifice and a cross flow velocity of 60 cm/s

From the results presented above, we can see that the bubble size varies fairly linearly with applied gas pressure. This trend agrees with Doku's findings. We can also conclude that the effect of varying gas pressure is similar for all four orifice diameters considered. From these figures it can be seen that bubble size decreases as the pressure decreases and therefore in order to generate microbubbles we only need to decrease the gas pressure the corresponding amount. However the gas pressure can only be decreased until the capillary pressure of the orifice is reached. At this point bubble generation ceases as the gas pressure can not overcome the capillary pressure. This is the reason that there are no data points below 60 psi for the 0.5 μm orifice. For pure water, the capillary pressure for

the 0.5 μm orifice is about 42 psi. Near this threshold pressure, bubble generation can be unstable and no data was taken at 45 psi.

5.4.3 Effect of Orifice Diameter on Bubble Size

To compare the results of bubbles generated at various orifice diameters we may plot bubble size versus cross flow velocity or pressure for various orifice diameters at a constant surfactant concentration. In order to present the trends observed in this work, this section will include those plots for a surfactant concentration of 0.01 % w/v. Figure 5-44 shows the results for bubble size versus pressure at a cross flow velocity of 60 cm/s while Figure 5-45 shows bubble size versus cross flow velocity at a pressure of 60 psi.

Figure 5-44 Bubble size versus pressure for various orifice diameters for a cross flow velocity of 60 cm/s and a surfactant concentration of 0.01 % w/v

Figure 5-45 Bubble size versus pressure for various orifice diameters for a pressure of 60 psi and a surfactant concentration of 0.01 % w/v

The figures above suggest that bubble size varies fairly linearly with orifice diameter for the larger orifice diameters. The effect of decreasing the orifice diameter appears to be slightly less pronounced between the 1 μm and 0.5 μm orifices. Figure 5-46 shows bubble size versus orifice diameter for a cross flow velocity of 60 cm/s and a surfactant concentration of 0.01% w/v for several gas pressures. From this figure we can see that there is a slight change in the slope between the 1 μm orifice and the 0.5 μm orifice. Unlike constant pressure bubble generation in a static liquid, the bubble size does not vary with the cube root of the orifice diameter. This is due to the fact that the buoyancy force is no longer a dominant detaching force. The primary detaching force is the drag force on the bubble due to the liquid cross flow.

Figure 5-46 Bubble size versus orifice diameter for a cross flow velocity of 60 cm/s, a surfactant concentration of 0.01 % w/v and various pressures

If we consider the two dominant forces to be the drag force on the bubble balanced by the surface tension force, we would expect a linear trend. By combining Equations 2-1, 2-2 and 2-6, noting that W is replaced by the liquid cross flow velocity, we get a linear relationship for bubble size with respect to orifice diameter which is shown below in Equation 5-3.

$$D_b = CD_0 \quad (5-3)$$

In Equation 5-3 C is a constant which is a function of the various physical parameters of the experiment. These include surface tension, cross flow velocity and liquid viscosity. Figure 5-47 shows the expected values for bubble size using Equation 5-3 and the data in Figure 5-46 with a gas pressure of 60 psi.

Figure 5-47 Expected trend compared to experimental results for bubble size versus orifice diameter for a cross flow velocity of 60 cm/s

As we can see from the figure above, the experimental trend is similar to the expected trend except at the smallest orifice diameter. The slopes of the trends are also slightly different with the experimental variation being less dependent on orifice diameter than we would expect. The surface tension force reduction reported by Nahra could be responsible for part of the difference in magnitude of the bubble sizes. In addition to this, the small contributions from the gas momentum and buoyancy could also reduce the size of the generated bubbles slightly. This argument is strengthened by the fact that the experimental results approach the expected trend for high cross flow velocities because the higher the flow velocity, the higher the drag force on the bubble. Figure 5-48 shows the experimental results versus the expected trend for the parameters in Figure 5-47 at a

flow velocity of 80 cm/s while Figure 5-49 shows the results for a flow velocity of 140 cm/s.

Figure 5-48 Expected trend compared to experimental results for bubble size versus orifice diameter for a cross flow velocity of 80 cm/s

Figure 5-49 Expected trend compared to experimental results for bubble size versus orifice diameter for a cross flow velocity of 140 cm/s

It should be noted that the experimental bubble size changes for different gas pressures as this is not taken into account by balancing the drag force with the surface tension force. It should also be noted that the 0% w/v data at a 5 μm does not agree with the surface tension trends because the pipette employed in the experiment cracked between experiments and was replaced. The replacement pipette is believed to have had a larger inner diameter.

Chapter 6: Formulation of a Theoretical Model to Predict Bubble Size Generated at Constant Pressure at a Micro-scale Orifice in a Static Liquid

A computer model to predict the diameter of bubbles generated from a given orifice at a given pressure and surfactant concentration is a useful tool. If a TBI bubble generator is to be designed at Artenga, it will be useful to know what size orifice is required given a required bubble diameter. In addition to being a useful tool, the model will yield insight into the physical phenomena of bubble generation. This model will also attempt to take the compressibility of the gas and the subsequent bubble expansion into account.

There are several theoretical models in literature which have been formulated to predict the detachment size of a bubble being generated at an orifice. These models have been compared to experimental data for a given range of orifice size and most show acceptable agreement. In all cases there are several common assumptions which are valid for large orifice sizes but are not necessarily valid for a micro-scale orifice. These include:

1. Gas is incompressible
2. The effect of the previous bubble's wake is negligible
3. The liquid remains static during the generation of each bubble
4. The contact angle between the bubble and the orifice is 0°

Since the capillary pressure at a micro-scale orifice can be quite large, the applied gas pressure required to generate a bubble can also be quite large. Since the applied gas pressure is high and the pressure inside the bubble is low, a situation involving compressible flow arises. In many cases the flow is choked and therefore the velocity and hence the volume flow rate is a constant. In this case, the mass flow rate must be considered rather than the volume flow rate. In addition to this change, the gas inside the bubble expands from the pressure at the orifice to atmospheric pressure. This expansion is an important factor that affects the forces acting on a bubble.

Although the previous bubble's wake can often be neglected, this is not the case for high frequency bubble generation at a micro-scale orifice. For constant pressure bubble generation, the pressure required to generate bubbles dictates that the bubble generation frequency will be relatively high. In such cases, the bubbles are close together and hence their wakes will affect the next bubble. For the experiments in this work, the combined effect of the wakes of many bubbles increases the local liquid velocity near the orifice such that the assumption of a static liquid medium is also invalid. The liquid is entrained in the bubble stream and a liquid flow in the direction of the rising bubble stream develops. This liquid flow tends to lift the forming bubble and detach it from the orifice causing the bubble to detach sooner compared to a bubble generated in a static liquid.

Finally, the contact angle between the bubble and the orifice is almost always assumed to be equal to 0° . This assumption is valid for orifice sizes as small as $5\text{ }\mu\text{m}$ however it fails below this size. Results have been presented in this work which demonstrate that the surface tension force is significantly reduced because the contact

angle at small orifice sizes can be large. This reduction is not negligible as the surface tension force is a dominant attaching force. As a result bubbles detach sooner than would be expected if the contact angle remained at 0° .

In this chapter a theoretical model for predicting the size of a bubble generated at constant applied gas pressure at a micro-scale orifice is derived. By considering a force balance on a bubble at the moment it detaches from the orifice, the bubble size is predicted. The model will incorporate the effects of the previously neglected components mentioned above. Following a derivation of the model, an overview of the solution technique which employs MATLAB will be described. Next the model results will be compared with the experimental results from section 5.2. The chapter will be concluded with a discussion of the results and an analysis of the validity of the model.

6.1 Model Derivation

In order to predict the detachment diameter of a bubble using a force balance, each force acting on a bubble at detachment must be known and a detachment criterion must be defined. In this work, a bubble is assumed to detach when the attaching forces are equal to the detaching forces. Although it is generally accepted that a bubble remains attached for a short time after the detaching forces become greater than the attaching forces, it is believed that under the conditions of the experiments in this work, it is valid to assume that this period will have a very small effect on final bubble size. This assumption is made because the size of the neck and the time the bubble rises after the detachment criteria has been met is very small for bubbles which have a diameter on the order of 100

times larger than the orifice. The validity of this “single stage model” for a small orifice was demonstrated by Goldschmidt et al [19].

By considering a single bubble just before detachment, we can conceive a number of forces which either act to keep a bubble attached to the orifice or promote detachment from the orifice. Any force which acts in the direction of gravity is considered an attaching force and any force which acts in the opposite direction is considered a detaching force. The attaching forces are:

1. Surface tension, F_σ
2. Inertia of the liquid that the bubble displaces as it forms, F_i
3. Drag on the bubble due to friction with liquid molecules, F_D

The detaching forces are:

1. Buoyancy of the bubble, F_B
2. Lift force due to the wake of the previous bubble and the liquid entrained by the bubble stream, F_L
3. Force due to the change in gas momentum, F_m

6.1.1 Dynamics of Bubble Formation

In order to model the formation of a bubble, the dynamics of the bubble formation must be known. For example, the velocity at which the bubble rises during formation must be known to calculate the drag force. The acceleration of the bubble must also be known in order to calculate the inertia force. Before the velocity and acceleration of the bubble during formation may be known, the gas flow rate into the bubble must be calculated. For high frequency bubble generation, the liquid medium will not be still, therefore this must

be taken into account by considering the effect of previous bubbles on the liquid medium. Finally, in order to calculate the wake effect, the velocity of the previous rising bubble at the time of detachment of the next bubble must be known.

Calculation of Gas Flow Rate

Before the gas flow rate through the orifice into the bubble can be calculated, we must consider the situation. Figure 6-1 shows a schematic of the gas flow path in this experiment.

Figure 6-1 Schematic of gas flow path showing gas pressures at various points

By considering basic fluid mechanics we know that the gas mass flow rate will be proportional to the pressure difference between the applied gas pressure at the exit of the orifice and the pressure inside the bubble. The gas velocity in the tube that connects the pipette to the reservoir is small because the pipette diameter is much smaller than the

tubing diameter. Therefore the gas pressure at the entrance to the tip of the pipette can be considered to be equal to the reservoir pressure.

The pressure inside the bubble will be a sum of the pressure head due to the liquid above the bubble, the pressure due to surface tension and atmospheric pressure. The pressure due to the bubble being submerged in the liquid is given by Equation 6-1 while the pressure due to surface tension is given by Equation 6-2.

$$p_h = \rho_w gh \quad (6-1)$$

$$p_\sigma = \frac{2\sigma}{D_b} \quad (6-2)$$

Combining these relations with the pressure due to atmospheric pressure, we obtain Equation 6-3.

$$p_b = p_{atm} + \rho gh + \frac{2\sigma}{D_b} \quad (6-3)$$

If we consider the magnitude of each term for the size of the generated bubbles relative to atmospheric pressure, we conclude that the pressure inside the bubble is nearly atmospheric. The pressure head due to the bubble being submerged is equal to 1” of water. At this depth, the pressure component is equal to 248.8 Pa. Compared to atmospheric pressure, this is equal to a change of 0.25% and is therefore negligible. Since no bubbles under 120 μm in diameter were generated, we can consider a bubble diameter of 100 μm to evaluate the maximum pressure due to surface tension. At this size and the maximum surface tension of 0.0723 N/m, the pressure inside the bubble due to surface

tension is 5,784 Pa or 5.7% of atmospheric pressure. Since the bubbles generated are larger than this value, it can be argued that the time average pressure inside a bubble during generation is approximately equal to atmospheric pressure.

Since the pressure inside the bubble is known and the pressure near the tip of the pipette is known, we may consider the pressure ratio between the tip of the pipette and the inside of the bubble to determine the gas flow characteristics. As mentioned previously, the pressure required to overcome the capillary pressure of the small pipettes is generally greater than 30 psi and therefore that is the lowest pressure considered in the experiments. Equation 6-4 shows the ratio between this minimum pressure and the pressure inside the bubble.

$$p_r = \frac{p_{atm}}{p_{oa}} = \frac{14.7}{(30 + 14.7)} = 0.33 \quad (6-4)$$

From the equation above we can see that this pressure ratio is below the critical pressure ratio [16] and therefore the flow is choked and sonic conditions apply at the exit of the pipette for all test conditions.

This unique situation arises due to the fact that the capillary pressure of the small orifices prevents gas flow at lower pressures. Previous work in literature has been performed with large orifices which require low pressures to drive the gas flow. As a result, the work found in literature has not considered the effect of sonic conditions at the orifice. The current work in literature considers the volume flow rate of gas through the orifice however since the flow is choked in the current work, the mass flow rate of the

gas must be considered. A modified volume flow rate can be derived considering the expansion of the gas once it is inside the bubble.

The mass flow rate into the bubble is given by Equation 6-5. Using this equation and compressible flow relations, we can derive Equation 6-6.

$$\dot{m}_g = \rho_g v_g A_o \quad (6-5)$$

$$\dot{m}_g = \frac{0.6339}{4} \frac{p_o}{RT} \sqrt{\gamma RT} \pi D_o^2 \quad (6-6)$$

We may derive an equivalent volume flow rate assuming the gas expands to atmospheric pressure inside the bubble by considering Equation 6-7 to yield Equation 6-8.

$$Q = \frac{\dot{m}_g}{\rho_{atm}} \quad (6-7)$$

$$Q_{eq} = \frac{0.6339}{4} \frac{\rho_o}{\rho_{atm}} \sqrt{\gamma RT} \pi D_o^2 \quad (6-8)$$

Q_{eq} is the volume flow rate of gas at atmospheric pressure flowing through the pipette to form bubbles. In this work we assume that the gas expansion is an isentropic process as the gas expands quickly compared to the time required for significant heat transfer to occur. This can be represented in the model by the following:

$$\frac{\rho_o}{\rho_{atm}} = \left(\frac{p_o}{p_{atm}} \right)^{\frac{1}{\gamma}} \quad (6-9)$$

The expansion of a bubble of diameter D_b , containing gas at pressure p_o expanding to pressure p_{atm} is given by:

$$D_b = D_{bo} \left(\frac{p_o}{p_{atm}} \right)^{\frac{1}{3\gamma}} \quad (6-10)$$

Velocity and Acceleration of the Bubble during Formation

Consider a single bubble at a point in time, as depicted in Figure 6-2, at an orifice with diameter D_o . The mass flow rate of gas through the orifice into the bubble is $\dot{m}_g$, the mass of the bubble is m_b , the diameter of the bubble is D_b , and the velocity and acceleration of the bubble are W and γ_b respectively.

Figure 6-2 Schematic of a bubble during formation

First consider the mass of the gas inside the bubble, which is given by the volume of the bubble, V_b , multiplied by the density of the gas inside the bubble, ρ_{gb} .

$$m_b = \frac{\pi}{6} D_b^3 \rho_{gb} \quad (6-11)$$

The change in mass of the bubble per unit time, $\frac{dm_b}{dt}$, is equal to the mass flow rate of gas through the orifice, $\dot{m}_g$, into the bubble.

$$\frac{dm_b}{dt} = \frac{\pi}{2} D_b^2 W \rho_{gb} \quad (6-12)$$

Now the mass flow rate into the bubble is given by:

$$\dot{m}_g = \rho_g v_g A_o = \rho_g v_g \frac{\pi}{4} D_o^2 \quad (6-13)$$

By combining Equations 6-12 and 6-13, we get:

$$W = \frac{\rho_g v_g D_o^2}{2 \rho_{gb} D_b^2} \quad (6-14)$$

If we take a derivative of W with respect to time, we get the acceleration of the bubble, γ_b .

$$\gamma_b = - \left(\frac{\rho_g v_g D_o^2}{\rho_{gb} D_b^2} \right)^2 \frac{1}{2 D_b} \quad (6-15)$$

Equation of Motion of a Rising Bubble

The motion of a small bubble rising in a liquid medium is defined by the size of the bubble and the liquid properties. More specifically, a balance between the drag force, inertia force due to the liquid and the buoyancy force will allow the velocity at any time to be determined. The drag on a rising bubble is given by Equation 6-16, the inertia force is given by Equation 6-17 and the buoyancy force acting on a spherical rising bubble is given by Equation 6-18. The velocity at which the bubble rises is v_b , M is the mass of the water displaced by the bubble and the volume of the bubble is V_b .

$$F_D = \frac{1}{2} \rho_w v_b^2 A_b C_D \quad (6-16)$$

$$F_I = \frac{1}{2} M \frac{dv_b}{dt} \quad (6-17)$$

$$F_B = \rho_w V_b \quad (6-18)$$

By summing the forces we get:

$$\frac{dv_b}{dt} = 2g - \frac{111 \rho_w^{\frac{3}{5}}}{4 \mu_w^{\frac{3}{5}}} D_b^{-\frac{8}{5}} v_b^{\frac{7}{5}} \quad (6-19)$$

Equation 6-19 is a nonlinear differential equation which can be solved numerically to obtain the velocity of the rising bubble, v_b , at a given time. The initial condition, that is, the bubble velocity at time zero, is equal to the velocity at which the bubble is rising during formation at the time of detachment.

When we compare the prediction of Equation 6-19 to experimental results from Graham Wallis' work [38], we see that the terminal velocity is under predicted. By modifying the drag coefficient, we obtain Equation 6-20 which agrees with experiment.

$$\frac{dv_b}{dt} = 2g - 0.38 \frac{111\rho^{\frac{3}{5}}}{4\mu_b^{\frac{3}{5}}} D_b^{-\frac{8}{5}} v_b^{\frac{7}{5}} \quad (6-20)$$

6.1.2 Attaching Forces

Surface Tension Force

The surface tension force is a direct connection between the bubble and the orifice which holds the bubble in place. The force is proportional to the surface tension of the liquid, the perimeter of the connection between the orifice and the bubble and the contact angle between the orifice and the bubble. The perimeter of the connection between the orifice and the bubble is not necessarily the internal perimeter of the orifice and could be some effective value between the outside diameter and the inside diameter, D_{eff} . Considering Figure 5-14 and Equation 2-1, the surface tension force is derived to give Equation 6-21.

$$F_\sigma = \pi D_{eff} \sigma \cos(\theta) \quad (6-21)$$

The value of σ is dependent on the bubble generation frequency. If the frequency is very high, then the time in which the bubble surface forms is small. If the time is much smaller than the time required for surfactant molecules to migrate to the gas-liquid interface, the surface tension during formation will be higher than the equilibrium surface

tension. Figure 4-9 shows the relation between dynamic surface tension and time. Since the bubble generation frequency for the 0.5 μm and 1 μm pipettes are less than 50 bubbles/s, the equilibrium surface tension values are used in the model. The 2 μm and 5 μm pipettes produce in excess of 500 bubbles/s, therefore the greatest dynamic surface tension values are employed.

Inertia Force

The inertia force or added mass force takes the inertia of the liquid that the bubble must accelerate into account. From the literature review, several sources have determined that this can be represented by Equation 2-14, where γ_b is the average acceleration of the bubble.

$$F_I = \left(\frac{\rho_{gb}}{\rho_w} + \alpha \right) \rho_w V \gamma_b \quad (2-14)$$

By substituting Equation 6-15 into Equation 2-14 and writing the volume of the bubble in terms of diameter, we get:

$$F_I = \frac{\pi}{12} \left(\frac{\rho_{gb}}{\rho_w} + \alpha \right) \left(\frac{\rho_g v_g D_o^2}{\rho_{gb} D_b} \right)^2 \quad (6-22)$$

Drag Force

The drag force on the bubble which opposes detachment is due to the viscous drag on the surface of the bubble as it grows. In order to be consistent with literature, we assume that the drag force acts on the entire surface area of the sphere. If this is the case, we can let the drag force be represented by Equation 6-23, where C_D is the drag coefficient given by

Equation 6-24. The relation for drag coefficient was chosen because Goldschmidt et al. [19] argued that this relation was more applicable for small bubbles than the Stokes drag relation.

$$F_D = D_{\text{mod}} \frac{1}{2} \rho_w W^2 S_b C_D \quad (6-23)$$

$$C_D = \frac{18.5}{\text{Re}_b^{\frac{3}{5}}} \quad (6-24)$$

$$\text{Re}_b = \frac{\rho_w W D_b}{\mu_w} \quad (6-25)$$

By combining Equations 6-23, 6-24 and 6-25, we get:

$$F_D = D_{\text{mod}} \frac{37\pi}{4} \rho_w^{\frac{2}{5}} D_b^{\frac{7}{5}} W^{\frac{7}{5}} \quad (6-26)$$

6.1.3 Detaching Forces

Buoyancy Force

The force on the bubble due to buoyancy is merely due to the difference in density between the gas in the bubble and the density of the liquid bulk. Assuming a spherical bubble, the difference in density multiplied by the volume of the bubble and the gravitational constant gives Equation 6-27.

$$F_b = g(\rho_w - \rho_{gb}) \frac{\pi}{6} D_b^3 \quad (6-27)$$

It is a common and reasonable assumption to consider the gas density to be negligible so that the relation becomes:

$$F_b = g\rho_w \frac{\pi}{6} D_b^3 \quad (6-28)$$

Lift Force due to Previous Bubble's Wake

According to Goldschmidt's application of Schlichting's boundary layer theory to flow around a sphere, the velocity defect behind a rising bubble is proportional to the velocity of the rising bubble to the power -2/3 [19,37]. They proposed the relation shown in Equation 6-29.

$$v_w = kv_b \left(\frac{\int_0^{\frac{1}{f}} v_b dt}{d_b} \right)^{-\frac{2}{3}} \quad (6-29)$$

In the expression above, the velocity of the bubble is integrated with respect to time over the time it takes to generate one bubble. This yields the distance that the bubble has travelled since it detached from the orifice and can be calculated through numerical integration of the solution of the equation of motion for a bubble rising in a liquid. The constant k is assumed to be 1.

From the relations above we know the velocity of the liquid at the orifice due to the previous bubble. This liquid velocity will induce a lift (detaching drag) force which will have the same form as the drag force discussed earlier however it acts in the opposite direction. This can be modelled by combining the drag force and lift force to form one

term which we will call the net drag force. The lift force is shown in Equation 6-30 and the net drag force is shown in Equation 6-31.

$$F_L = D_{\text{mod}} \frac{37\pi}{4} \rho_w^{\frac{2}{5}} D_b^{\frac{7}{5}} v_w^{\frac{7}{5}} \quad (6-30)$$

$$F_{D_{\text{net}}} = \frac{37\pi}{4} D_{\text{mod}} \rho_w^{\frac{2}{5}} D_b^{\frac{7}{5}} (v_w - W) |v_w - W|^{\frac{2}{5}} \quad (6-31)$$

We can see from Equation 6-31 that whenever the wake velocity is greater than W , the net drag force will be a detaching force.

In the present work, the bubble generation frequency is high. If we assume that each bubble affects the one after it, we can argue that if the generation frequency increases, the velocity of the rising bubble stream will also increase. This is because the fluid behind a rising bubble has a non-zero velocity. As the next bubble passes through this region, the absolute velocity at which it rises will increase. This has been demonstrated in literature for pairs of bubbles and it has been shown that the next bubble will often catch up to the previous bubble [21,39]. By considering this evidence, we can reason that the lift force will be increased by this phenomenon and therefore a drag modifier, D_{mod} , has been added to take this into account. The factor D_{mod} will also account for any liquid bulk recirculation effects. We believe that this effect is insignificant as experiments have revealed that the liquid entrainment velocity due to the bubble stream is far greater than the liquid bulk recirculation velocity.

Force due to Change in Gas Momentum

The force due to the change in gas momentum can be calculated by calculating the gas momentum at the orifice exit and the gas momentum inside the bubble. In this work, the gas velocity at the orifice is constant and equal to the speed of sound at room temperature, v_o . The density of the gas at the exit can be calculated using compressible flow relations [16]. If we assume that the gas velocity inside the bubble is equal to the velocity of the bubble, the force due to the change in momentum is given by:

$$F_m = \dot{m}_o v_o - \dot{m}_o W = \dot{m}_o (v_o - W) \quad (6-32)$$

We know that v_o is much greater than W , therefore:

$$F_m = \dot{m}_o v_o \quad (6-33)$$

$$F_m = \rho_o v_o^2 A_o \quad (6-34)$$

This expression is accepted in literature and employed in many models. Generally the term for gas momentum is considered to be negligible because it is small compared to other forces. In our case, the magnitude of the force given by Equation 6-34 is on the same order of magnitude as the other forces in our force balance. In literature, the ratio of bubble diameter to orifice diameter is usually less than 10. In this work the ratio is as large as 400. This large ratio suggests that there could be a large amount of recirculation within the bubble. In addition to this, the gas undergoes a large sudden expansion which will induce flow losses and reduce the gas momentum force.

If we assume that the expansion from the orifice to the bubble can be represented by a pipe flow with a sudden expansion, we know that for a pipe flow undergoing a sudden expansion, the loss when the area of the expansion is much larger than the area of the pipe is equal to the velocity head. By considering Bernoulli's equation from a point at the orifice to a point in the centre of the bubble, we can see that the velocity inside the bubble is small due to the large loss. If we further consider the recirculation of gas within the bubble due to the fact that the bubble diameter is much greater than the orifice diameter, it is reasonable to assume that the change in gas momentum due to the bubble wall is very small. Since we expect the gas momentum force to be much smaller than Equation 6-34 suggests, a reduction factor, R_m , is introduced to yield:

$$F_m = R_m (\rho_o v_o^2 A_o) \quad (6-35)$$

By comparing model results with experimental results, R_m is assigned a value of 0.1.

6.1.4 Constitutive Equation

As mentioned in the previous section, the model in this work assumes that a bubble will detach when the detaching forces equal the attaching forces. To obtain the constitutive equation in which all terms are known except bubble diameter, we set the expressions for detaching forces equal to the expressions for attaching forces to get:

$$F_b + F_{Dnet} + F_m = F_\sigma + F_I \quad (6-36)$$

$$2\rho_w D_b^3 + 111D_{\text{mod}} \rho_w^{\frac{2}{5}} D_b^{\frac{7}{5}} \left(v_w - \frac{\rho_o v_o D_o^2}{2\rho_{gb} D_b^2} \right) \left| v_w - \frac{\rho_o v_o D_o^2}{2\rho_{gb} D_b^2} \right|^{\frac{2}{5}} = 12D_{\text{eff}} \sigma \cos(\theta) + \left(\frac{\rho_{gb}}{\rho_w} + \alpha \right) \left(\frac{\rho_g v_g D_o^2}{\rho_{gb} D_b} \right)^2$$

(6-37)

6.2 Solution Technique

To obtain the value of bubble diameter which satisfies Equation 6-37 for a given set of parameters, a root finding algorithm *fsolve*, included in MATLAB, is used. As the wake velocity, v_w , is a function of bubble diameter, the process must be iterative. To obtain v_w we must integrate the numerical solution of the equation of motion of a bubble rising in a liquid. Therefore Equation 6-37 is first solved assuming v_w is 0. The value for D_b obtained is then used to calculate v_w . Once v_w is known, the equation is solved again to find a new D_b . This process is repeated until the value of D_b changes by less than 1%. A flow chart depicting the solution method is shown in Figure 6-3. Appendix B includes the MATLAB code employed and the values of the physical parameters.

Figure 6-3 Flow chart depicting model solution method

6.3 Model Results

The constitutive equation was solved for the various sets of physical parameters for which the static constant pressure bubble generation experiments were performed. The results for bubble diameter versus applied gas pressure are shown in Figure 6-4 through Figure 6-7. The experimental data is represented as points while the model predictions are shown as solid lines. Bubble diameter decreases as surfactant concentration increases for the model predictions.

Figure 6-4 Predicted bubble size compared to experimental data for a 0.5 μm orifice

Figure 6-5 Predicted bubble size compared to experimental data for a 1 μm orifice

Figure 6-6 Predicted bubble size compared to experimental data for a 2 μm orifice

Figure 6-7 Predicted bubble size compared to experimental data for a 5 μm orifice

From the figures above we can see that there is good agreement between the model and experimental data. The greatest disagreement between the model and the experimental data is approximately 13%. We see that this disagreement occurs for the 0.5 μm data. The model underestimates the change in bubble size with pressure. The model also appears to over predict the bubble size for the 1 μm data at low pressures. The sensitivity to changes in surface tension also exhibits a slight disagreement for all cases. There are several possible explanations for the discrepancies which exist.

It has been argued in literature that the smaller the orifice becomes, the more sensitive the bubble size is to changes in orifice geometry. In this work we can see a greater disagreement for the smaller orifice sizes. It is possible that small variations in the orifice geometry play a role in the results. For example, an orifice which has a circular cross

section may have a 3-dimensional variation which increases the perimeter of the orifice, and hence the surface tension force. This variation could also cause the gas to flow asymmetrically. These effects are not accounted for in the model.

For the 5 μm orifice, the bubble diameter is slightly more sensitive to applied gas pressure than the experimental data suggests it should be. This is likely due to the assumption that the gas momentum is 10% of the calculated value. For the smaller orifice diameters, the ratio of bubble diameter to orifice diameter is usually greater than 150. For the 5 μm orifice, this ratio is as small as 60. Although this is still quite large, it is likely that the gas momentum force is reduced by a smaller factor. This variation could be taken into account if a relation was derived for the losses inside an expanding or forming bubble.

Alternatively, it is possible that the gas momentum force is negligible for the 5 μm orifice and another factor is responsible for the disparity. In this work it is assumed that the drag increase at high bubble generation frequencies is the same for each orifice. In fact, it is likely that the increase varies with orifice diameter. This argument is based on the fact that the bubble diameter, rise velocity and generation frequency increase with orifice diameter. A larger bubble stream which contains a greater number of bubbles rising at a greater velocity will undoubtedly entrain the liquid bulk more effectively. This would result in a greater lift force on the bubbles which would promote detachment. This argument is strengthened when we consider the fact that bubble generation frequency increases with pressure.

By this reasoning, we see that the results suggest that the drag force for the 0.5 μm orifice results is slightly larger than it should be while for the 5 μm orifice, the drag force is slightly smaller than it should be.

The current model could be improved by modeling the bubble stream as a cylinder of varying density rising through the liquid. The density of the column would be a function of the generation frequency. The rise velocity of the bubble stream and the entrained liquid could therefore be written as a function of the column density. This approach would allow the variations in the drag force described above to be taken into account.

Another area for improvement is the relation between orifice contact angle and orifice diameter. In the current work, empirical data is used to set the value of the contact angle in the model. A method for predicting the contact angle for any combination of orifice size, material and liquid surface tension would strengthen the model significantly from a theoretical perspective. To predict the contact angle for orifice diameters similar to the pipettes employed in this work, a curve could be fit to the empirical values.

The surface tension values used in this model could be improved by creating a function which represents Figure 4-9. Given the bubble generation frequency, the model could call the function and receive the proper surface tension value for the given conditions. This feature would likely improve the model's response to changes in surface tension as the model would no longer assume either the largest dynamic surface tension value or the equilibrium value.

The model derived in this work is satisfactory as a tool to predict the generated bubble size for a constant gas pressure bubble generator given the orifice diameter, gas

pressure and surfactant concentration. The size of bubbles generated at a choked orifice is successfully predicted and the model provides insight into the physical parameters involved in bubble generation.

Chapter 7: Feasibility of Microbubble Generator use in the Artenga TBI

There are two main requirements that must be met by a microbubble generator if it is to be used in the TBI. These requirements are that the bubble diameter should be between 1-5 μm and the concentration of bubbles should be about 10^6 bubbles per millilitre. By looking directly at the experimental data, it seems that the size range cannot be met by the current technique and the concentration requirement can be met granted there is time to allow the bubbles to accumulate. During work done developing a two phase flow generator at Artenga, there were some interesting observations which that were made that impact the decision as to whether this device is feasible or not. The first observation was that small bubbles are unstable and dissolve quickly. The second was that the bubbles could be stabilized using a binary system of surfactants. Lastly, during the development of a two-phase flow device, contact with various researchers identified advantages to the constant pressure bubble generation techniques.

7.1 Bubble Instability

Work on a two phase flow bubble generator was done in parallel to the work concerned in this thesis. The two phase approach involved forcing liquid and gas to flow

simultaneously through a capillary tube having a diameter of about 200 μm . The technique is depicted in Figure 7-1.

Figure 7-1 Two phase flow approach to generating microbubbles

The process uses two syringes connected by a capillary tube. Initially, one syringe contains liquid and a small amount of gas headspace. By passing the mixture through the capillary tube into the other syringe, many small bubbles are formed. After some optimization, we determined that the technique could generate bubbles which met the size requirements and exceeded the concentration requirements initially outlined. There was a problem however, which was not foreseen. We observed that bubbles generated in a solution containing a single surfactant were unstable. Any bubbles below the size of 20 μm quickly disappeared. The effect was very dramatic for bubbles under 10 μm , as they persisted for only a few seconds. The conclusion that was drawn was small bubbles that met the requirements would quickly disappear. Figure 7-2 shows the behaviour of bubbles comprised of various gases.

Figure 7-2 Bubble dissolution in water for bubble comprised of several different gases

This result suggests that although bubbles from the constant pressure bubble generating system are larger than those required for the TBI, the bubbles could be allowed to shrink over time and hence meet the requirements. The constant pressure bubble generation in a static liquid technique yields bubbles which are too large for this technique to be viable. The smallest bubble generated in a static liquid at constant gas pressure is over 100 μm in diameter. The time required for a 100 μm bubble to shrink is too long for this approach to be practical. This is not necessarily true for constant gas pressure bubble generation in a cross flow. For high liquid cross flow velocities and a small orifice, it is possible to generate bubbles which are 20 μm in diameter. A 20 micron bubble will only require about 10 – 20 seconds to become less than 5 μm in diameter.

It is evident from Figure 7-2 that the bubbles are unstable and shrink until they disappear. Without a method to stabilize the microbubbles, they will dissolve within a few seconds. This problem is solved by using a binary surfactant system consisting of a soluble surfactant and a solid insoluble surfactant. The soluble surfactant allows the small bubbles to be formed, while the insoluble surfactant, which is in suspension in the liquid, forms an insoluble shell on the bubble. As the bubble shrinks, the soluble surfactant goes into solution and is replaced by insoluble surfactant until the bubble is stabilized. Using this technique, microbubbles can be stabilized for several hours or more.

7.2 Bubble Concentration

Although the microbubbles can be stabilized at the target size if we use the cross flow technique, we must also consider the bubble concentration requirement. For an orifice in the size and pressure range considered in this work, it is reasonable to assume the bubble generation frequency is between 100 and 500 bubbles per second. Even if there was only 1 ml of liquid medium in which the bubbles were being generated, it would take over a day to generate 1 ml of microbubbles at the target concentration. This is assuming that the bubbles are stable for an entire day. Even under the assumption that the bubbles would be stable for several days, this time estimate is far too large for use in an on-demand device. In a cross flow system it is unlikely that the liquid medium could be reduced to much less than a 200 ml. In order for this system to feasibly meet the requirements set out at the outset of the project, a system with many orifices would have to be designed. It may be possible to achieve such a design with MEMS technology,

however from a cost and simplicity of design perspective, the two phase flow system is much more feasible.

7.3 Cross Flow Advantages and Applications

Although the system does not meet the requirements set out at the beginning of the project, there are several advantages to the cross flow technique over other techniques which are currently available. These are:

1. Homogeneity
2. Control over bubble size
3. Ability to directly observe generation

In the field of microbubbles and ultrasound, many researchers have expressed interest in obtaining microbubbles which are homogenous in size. That is, the variation in size among a bubble population is small. Current generation techniques, including the Artenga two phase flow approach, generate a population of bubbles which are distributed around a mean size with, for example, 90% of bubbles with a diameter between 1 μm – 5 μm . Using the cross flow technique, the bubble size variation would be much smaller than this.

In addition to the ability to generate homogeneous bubbles, the techniques described in this work allow the bubble size to be varied simply by adjusting gas pressure or liquid cross flow velocity. Several researchers have expressed interest in performing research with homogeneous microbubbles which are larger than the microbubbles which are currently available [40]. In some cases, a specialized bubble shell [40] would be required and further work may be required to characterize bubble generation in various fluids. An

example of a research application is the studies geared towards studying single bubble, cell and ultrasound interaction performed by Van Wamel et al. [9].

Another advantage of the cross flow technique is that bubble generation can be observed using the apparatus designed in this work. If the properties of the liquid changed significantly, for example if the viscosity was increased greatly, the mechanisms involved in bubble generation may vary. Direct observation of the bubble neck and shape could yield insight into these mechanisms.

With some development, the cross flow technique could be used to generate homogeneous bubbles of a specified size at low concentration. For the current TBI, the two phase flow technique provides a method to generate microbubbles to meet the requirements. Artenga will continue to develop the TBI device and perform in vitro and in vivo testing to determine the optimum bubble and ultrasound parameters which will lead to a treatment plan for drug resistant tumours.

Chapter 8: Conclusions and Recommendations

Although the bubble generation techniques employed in the experiments in this work will likely not be used in the Artenga TBI device, the techniques will be applicable to future device development and research work. Several researchers have expressed interest in the capability of producing homogeneous bubbles of a specific diameter. In addition to this, the current work provides data and insight into the field of bubble generation at a micro-scale orifice which did not previously exist. The effects of surface tension reduction through the use of surfactants, the use of high pressure compressible gas and liquid cross-flow on bubble generation at a micro-scale orifice have been investigated. This work has demonstrated that the bubble wake effect is an important and sometimes dominant effect for high frequency bubble generation at a micro-scale orifice. The theoretical model is a validation of the expression proposed by Goldschmidt et al. to describe the velocity defect behind a rising bubble.

8.1 Recommendations

Although the current model is an acceptable tool for predicting bubble size given the conditions of bubble generation, the model could be improved by considering the following changes. By modeling the rising bubble stream as a cylinder of varying density rising through the liquid, a more representative liquid bulk velocity could be obtained.

The density of the column would depend on the generation frequency. The rise velocity of the bubble stream and hence the entrained liquid could therefore be written as a function of the column density. This approach would allow the variations in the drag force described in the model results section to be taken into account.

Another area for improvement is the relation between orifice contact angle and orifice diameter. In the current work, empirical data is used to set the value of the contact angle in the model. A method for predicting the contact angle for any combination of orifice size, material and liquid surface tension would strengthen the model significantly from a theoretical perspective. To predict the contact angle for orifice diameters similar to the pipettes employed in this work, a curve fit of empirical data could be used.

The surface tension values used in this model could be improved by creating a function which represents Figure 4-9. Given the bubble generation frequency, the model could call the function and receive the proper surface tension value for the given conditions. This feature would likely improve the model's response to changes in surface tension as the model would no longer assume either the largest dynamic surface tension value or the equilibrium value.

In order to produce a cross flow bubble generation device that would generate acceptable concentrations of microbubbles, the liquid in the flow loop must be minimized. This could be done by manufacturing a smaller flow channel, tubing loop and incorporating a miniature pump and reservoir system. By adding multiple orifices at which bubbles could be generated, the system could conceivably become a system which

could generate homogeneous bubbles at moderate concentrations. This would likely be best achieved using a custom MEMS device.

Significant theoretical work can be continued towards modelling the bubble wake effect. Data gathered during the constant flow experiments is not fully processed and could yield interesting results. By expanding on the frame by frame analysis technique employed in this work, we could calculate the velocity of each tracked particle and the distance from the bubble. By tracking many particles, we could obtain a velocity distribution of the liquid bulk behind a rising bubble and determine whether the velocity defect in fact varies with the distance from the bubble to the $-2/3^{\text{rd}}$ power. Careful inspection of the data and collection of new data using a greater concentration of Artenga microbubbles would yield interesting insights that could lead to a greater understanding of bubble generation at a micro-scale orifice.

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Appendix A: Complete Experimental Results

Constant Pressure Bubble Generation in a Liquid Cross Flow

Ex1

D = 1 micron

No surfactant

Bubble Size

	velocity	10	20	40	60	80	180	cm/s
	pressure (psi)			Bubble Diameter (μm)				
	30	137.5	102.5	62.5	42.5	37.5	20	μm
	45	220	162.5	75	62.5	42.5	27.5	μm
	60	262.5	175	95	67.5	47.5	30	μm
	75	275	185	100	75	55	35	μm
	90	312.5	250	115	85	65	37.5	μm

Ex2

D = 1 micron

C = 0.001% w/v

Bubble Size

	velocity	10	20	40	60	80	180	cm/s
	pressure (psi)			Bubble Diameter (μm)				
	30	167.5	85	62.5	42.5	35	17.5	μm
	45	185	140	85	55	42.5	22.5	μm
	60	220	147.5	90	65	52.5	27.5	μm
	75	250	180	110	75	65	32.5	μm
	90	317.5	202.5	132.5	90	65	37.5	μm

Ex3

D = 1 micron

C = 0.01% w/v

Bubble Size

velocity	10	20	40	60	80	180	cm/s
pressure (psi)	Bubble Diameter (μm)						
30	130	87.5	62.5	50	37.5	20	μm
45	137.5	112.5	80	60	47.5	26.25	μm
60	160	122.5	87.5	67.5	55	30	μm
75	200	140	110	77.5	60	35	μm
90	212.5	167.5	115	85	72.5	37.5	μm

Ex4

D = 1 micron

C = 1% w/v

Bubble Size

velocity	10	20	40	60	80	180	cm/s
pressure (psi)	Bubble Diameter (μm)						
30	77.5	50	37.5	30	27.5	20	μm
45	87.5	60	50	37.5	35	22.5	μm
60	95	70	57.5	42.5	37.5	25	μm
75	100	75	62.5	50	42.5	27.5	μm
90	105	90	77.5	60	52.5	30	μm

Ex 5

D = 2 micron

No surfactant

Bubble Size

velocity	10	20	40	60	80	150	cm/s
pressure (psi)	Bubble Diameter (μm)						
30	Too Big	350	192.5	117.5	100	67.5	μm
45	-	387.5	200	137.5	115	82.5	μm
60	-	460	205	142.5	127.5	97.5	μm
75	-	510	210	165	142.5	105	μm
90	-		222.5	172.5	157.5	110	μm

Ex 6

D = 2 micron

C = 0.001% w/v

Bubble Size

velocity	10	20	40	60	80	150	cm/s
pressure (psi)	Bubble Diameter (μm)						
30	Too Big	337.5	180	110	95	60	μm
45	-	375	185	130	110	80	μm

60	-	362.5	210	162.5	132.5	95	μm
75	-	387.5	215	167.5	140	105	μm
90	-		227.5	187.5	152.5	115	μm

Ex 7

D = 2 micron

C = 0.01% w/v

Bubble Size

velocity	10	20	40	60	80	150	cm/s
pressure (psi)	Bubble Diameter (μm)						
30	Too Big	262.5	167.5	115	95	62.5	μm
45	-	275	180	130	110	80	μm
60	-	305	195	152.5	127.5	87.5	μm
75	-	315	202.5	160	135	100	μm
90	-	325	222.5	180	147.5	105	μm

Ex 8

D = 2 micron

C = 1% w/v

Bubble Size

velocity	10	20	40	60	80	150	cm/s
pressure (psi)	Bubble Diameter (μm)						
30	Too Big	165	112.5	100	87.5	60	μm
45	-	187.5	142.5	115	102.5	77.5	μm
60	-	217.5	162.5	137.5	115	92.5	μm
75	-	245	180	157.5	130	105	μm
90	-	262.5	195	170	140	115	μm

Ex 9

D = 5 micron

No surfactant

Bubble Size

velocity	20	30	40	60	80	150	cm/s
pressure (psi)	Bubble Diameter (μm)						
30	375	0	270	227.5	212.5	170	μm
45	425	0	317.5	262.5	237.5	190	μm
60	0	407.5	377.5	327.5	282.5	207.5	μm
75	0	487.5	405	357.5	330	237.5	μm
90	0	0	437.5	385	362.5	262.5	μm

Ex 10

D = 5 micron

C = 0.001% w/v

Bubble Size

velocity pressure (psi)	30	40	60	80	150	cm/s
	Bubble Diameter (μm)					
30	437.5	400	375	350	262.5	μm
45	-	430	400	375	300	μm
60	-	475	450	420	337.5	μm
75	-	575	487.5	450	375	μm
90	-	-	comp	487.5	412.5	μm

Ex 11

D = 5 micron

C = 0.01% w/v

Bubble Size

velocity pressure (psi)	20	40	60	80	150	cm/s
	Bubble Diameter (μm)					
30	387.5	325	287.5	250	187.5	μm
45	417.5	362.5	325	287.5	225	μm
60	462.5	407.5	355	325	245	μm
75	507.5	450	387.5	332.5	262.5	μm
90	562.5	487.5	412.5	387.5	307.5	μm

Ex 12

D = 5 micron

C = 1% w/v

Bubble Size

velocity pressure (psi)	20	40	60	80	150	cm/s
	Bubble Diameter (μm)					
30	337.5	302.5	260	217.5	185	μm
45	365	327.5	287.5	250	215	μm
60	400	367.5	322.5	297.5	225	μm
75	435	400	355	312.5	260	μm
90	475	432.5	390	357.5	287.5	μm

Ex 13

D = 0.5 micron
No surfactant
Bubble Size

velocity	10	20	40	60	80	140	cm/s
pressure (psi)							
30	pressure too low						
45	Bubble Diameter (μm)						
60	127.5	68.75	56.25	40	35	23.75	μm
75	132.5	80	62.5	46.25	40	27.5	μm
90	137.5	87.5	70	50	43.75	30	μm

Ex 14
D = 0.5 micron
C = 0.001% w/v
Bubble Size

velocity	10	20	40	60	80	140	cm/s
pressure (psi)							
30	pressure too low						
45	Bubble Diameter (μm)						
60	122.5	60	50	37.5	33.75	23.75	μm
75	130	75	60	43.75	37.5	25	μm
90	132.5	85	66.25	47.5	41.25	27.5	μm

Ex 15
D = 0.5 micron
C = 0.01% w/v
Bubble Size

velocity	10	20	40	60	80	140	cm/s
pressure (psi)							
30	pressure too low						
45	Bubble Diameter (μm)						
60	107.5	67.5	60	47.5	40	27.5	μm
75	115	77.5	67.5	50	42.5	30	μm
90	125	90	72.5	55	45	32.5	μm

Ex 16
D = 0.5 micron
C = 1% w/v

Bubble Size

velocity pressure (psi)	10	20	40	60	80	140	cm/s
30	pressure too low						
45	Bubble Diameter (μm)						
60	62.5	42.5	37.5	32.5	27.5	20	μm
75	67.5	47.5	42.5	37.5	31.25	25	μm
90	72.5	52.5	47.5	40	33.75	28.75	μm

Constant Pressure Bubble Generation in a Static Liquid

	p (psi)	30	45	60	75	90
		Bubble Diameter (μm)				
0.5 (μm)	C=0			175	195	215
	0.001%			162.5	182.5	200
	0.01%			300	135	150
	1%			237.5	112.5	120
1 (μm)	C=0	232.5	267.5	305	320	325
	0.001%	250	287.5	325	337.5	342.5
	0.01%	362.5	207.5	227.5	242.5	245
	1%	142.5	185	200	225	232.5
2 (μm)	C=0	287.5	300	317.5	332.5	337.5
	0.001%	275	295	317.5	327.5	332.5
	0.01%	262.5	282.5	305	315	320
	1%	215	237.5	270	280	285
5 (μm)	C=0	362.5	382.5	402.5	425	437.5
	0.001%	350	372.5	392.5	407.5	422.5
	0.01%	337.5	360	380	395	407.5
	1%	292.5	330	362.5	382.5	395

Constant Gas Flow Bubble Generation in a Static Liquid

Do	Flow Rate ($\mu\text{l}/\text{min}$)	Db (μm)	f (bubbles/s)
1 (μm)	62	370	0.13
	94	361	0.15
	132	355	0.17

156	350	0.20
231	350	0.40
275	350	0.83
338	341	1.00
400	350	1.67

Do	Flow Rate ($\mu\text{l}/\text{min}$)	Db (μm)	f (bubbles/s)
2 (μm)	62	370	0.32
	94	361	0.36
	132	355	0.53
	156	350	0.69
	231	350	1.00
	275	350	1.67
	338	341	2.50
	400	350	4.00

Do	Flow Rate ($\mu\text{l}/\text{min}$)	Db (μm)	f (bubbles/s)
5 (μm)	62	370	0.12
	94	361	0.15
	132	355	0.36
	156	350	0.45
	231	350	0.67
	275	350	0.83
	338	341	> 5.00

Appendix B: MATLAB Code for Solution of Constitutive Equation

Bubbles.m

This is the master file which calls the other programs.

```
format long;
results=[];
resultsunformatted=[];

count=1;
countt=1;

bubblevar;

for l=1:4    % Loop varies orifice diameter

    Dnot=dnot(l);
    Deff=deff(l);
    A=.25*pi*Dnot^2;
    bubblevar;
    for m=1:4

        visc=viscmmod(countt);
        countt=countt+1;

        for n=1:5    % Loop varies pressure

            vw=0;
            sigmaa=sigma(count);    % count varies surface tension and contact angle for each data point
            angl=ang(count);
            count=count+1;

            pnot=p(n);

            rhog=.6339*(pnot+patm)/R/Tnot;

            options = optimset('TolFun',1e-40);

            dmod=Dmod(l);

            % next line solves for initial bubble diameter without wake velocity

            [x,fval]=fsolve(@constitutiveeq,200E-6,options,pnot,Dnot,Deff,vw,sigmaa,angl,rhog,dmod,visc);
            h=x*((.6339*(pnot+patm)/patm)^(1/(3*1.4)));

            rb=h;
```

```

rb1=2*rb;

while (rb1-rb)/rb1 > 0.01      % iterate until solution changes by less than 1%

    f=(6/4)*((.6339*(pnot+patm)/patm)^(.71))*vnot*(Dnot^2)/(rb^3);

    rb1=rb;

    W=(.5*((.6339*(pnot+patm)/patm)^(.71))*vnot*(Dnot^2)/(rb^2));

    stpsz=(1/f/12);
    options = odeset('MaxStep',stpsz);

    tmax=2/f;

    [t,y]=ode45(@bubblemotion,[0,tmax],[W],[options],rb); % solves for velocity of rising previous bubble
    vectorfix;

    q = length(tn);

    while q < 3

        stpsz=stpsz/12;
        options = odeset('MaxStep',stpsz);

        tmax = 1/f/100;
        [t,y]=ode45(@bubblemotion,[0,tmax],[W],[options],rb);
        vectorfix;
        q = length(tn);

    end

    inted=cumsimpson(tn,yn);

    distanc=inted(cont-1);

    vmod=Vmod(l);

    W2=yn(cont-1);

    vw=vmod*W2*(distanc/rb)^(-2/3);      % calculate wake velocity

    options = optimset('TolFun',1e-40,'TolX',1E-50,'MaxFunEval',1E10,'MaxIter',1E10);

    % next line solves for bubble diameter including wake velocity
    [x,fval] = fsolve(@constitutiveeq,200E-6,options,pnot,Dnot,Deff,vw,sigmaa,angl,rhog,dmod,visc);
    h=x*((.6339*(pnot+patm)/patm)^(1/(4.2))); % isentropic bubble expansion
    rb=h
    Dnot
end

hold all;

plot(tn,yn);

resultsunformatted=[resultsunformatted,h];

end

results=[results;,resultsunformatted];
resultsunformatted=[];

end

end
end

```

end

```
results
save testresults.txt results /ascii;
```

Bubblevar.m

This file contains the physical parameters and variables called by Bubbles.m

```
patm=101300;
k=1.4;
R=297;
Tnot=298;
rho1=998.3;
alph=1/2;
vnot=sqrt(k*R*Tnot);

p=[206734.6939
310102.0408
413469.3878
516836.7347
620204.0816
];

dnot=[.5E-6
.9E-6
3.25E-6
6.2E-6
];

deff=[.75E-6
1.65E-6
2.75E-6
6E-6
];

Dmod=[2 2 2 2];

Vmod=[1 1 1 1];

viscmod= [ .001
.001
.0015
.002
.001
.001
.0015
.002
.001
.001
.0015
.002
.001
.001
.0015
.002];

sgma=[72.3E-3
72.3E-3
72.3E-3
72.3E-3
72.3E-3
65E-3
```

65E-3
65E-3
65E-3
65E-3

52E-3
52E-3
52E-3
52E-3
52E-3

46E-3
46E-3
46E-3
46E-3
46E-3

72.3E-3
72.3E-3
72.3E-3
72.3E-3
72.3E-3

65E-3
65E-3
65E-3
65E-3
65E-3

52E-3
52E-3
52E-3
52E-3
52E-3

46E-3
46E-3
46E-3
46E-3
46E-3

72.3E-3
72.3E-3
72.3E-3
72.3E-3
72.3E-3

68E-3
68E-3
68E-3
68E-3
68E-3

63.5E-3
63.5E-3
63.5E-3
63.5E-3
63.5E-3

60E-3
60E-3
60E-3
60E-3
60E-3

72.3E-3
 72.3E-3
 72.3E-3
 72.3E-3
 72.3E-3

68E-3
 68E-3
 68E-3
 68E-3
 68E-3

63.5E-3
 63.5E-3
 63.5E-3
 63.5E-3
 63.5E-3

60E-3
 60E-3
 60E-3
 60E-3
 60E-3

];

% contact angle in radians. Note sin(90-ang) is used in the solution code. This is the way it was originally programmed and was not changed to be the same as the figures in the work above.

ang=[

.28
 .28
 .28
 .28
 .28

.28
 .28
 .28
 .28
 .28

.28
 .28
 .28
 .28
 .28

.28
 .28
 .28
 .28
 .28

.6
 .6
 .6
 .6
 .6

.6
 .6
 .6
 .6
 .6

bubblemotion.m

This file contains the function for the nonlinear differential equation of bubble motion.

```
function dydt = vdphil(t,y,d)

dydt = [2*9.81-(.38*18.5*3/2)*((d)^(-8/5))*((0.001/998.3)^(3/5))*(y^(7/5))];
```

constitutiveeq.m

This file contains the function for the constitutive equation. Attaching forces = detaching forces.

```
function F = constitutiveeq(x,pnot,Dnot,Deff,vw,sigmaa,angl,rhog,dmod,visc)

bubblevar;

F=[-dmod*(1/8)*(rho1^(2/5))*18.5*pi*(visc^(3/5))*((.5*((.6339*(pnot+patm)/patm)^(.71))*vnot*(Dnot^2)/(x^2)-vw)^(5/5))*abs((.5*((.6339*(pnot+patm)/patm)^(.71))*vnot*(Dnot^2)/(x^2)-vw)^(2/5))*(x^(7/5))-
(alpha+rhog/rhol)*(rhol/12)*((.6339^2)*(pnot+patm)/patm)^(2*.71))*vnot^2*(Dnot^4)*pi/(x^2)-
pi*Deff*sigmaa*sin(angl)+0*(pi/4)*(Dnot^2)*(.6339*pnot-patm-
sigmaa/x)+0.1*rhog*(vnot^2)*pi*(Dnot^2)/4+(pi/6)*rhol*9.81*(x^3)];
```

cumsimpson.m

This file contains a numerical integrator to integrate the numerical solution of the equation of motion.

```
function z=cumsimpson(x,y)
%CUMSIMPSON Cumulative Simpson Numerical Integration.
% Z=CUMSIMPSON(Y) computes an approximation of the cumulative integral of Y
% using a Simpson rule with unit spacing between the data points in Y. To
% compute the integral for spacing different from one, multiply Z by the
% spacing increment. When Y is a matrix, the cumulative integral is
% computed over each column of Y.
%
% Z=CUMSIMPSON(X,Y) computes the integral with respect to the data in
% vector X. X need NOT be EQUALLY spaced but must have the same number of
% elements as Y. When Y is a matrix, Y must have as many rows as X has
% elements.
%
% Z has the same dimensions as Y.
%
% See also CUMSUM, CUMTRAPZ, QUAD, QUADV.

% D.C. Hanselman, University of Maine, Orono, ME 04469
% MasteringMatlab@yahoo.com
% Mastering MATLAB 7
% 2005-09-30, 2006-02-21

if nargin<2
    y=x;
    [ry,cy]=size(y);
    if ry==1
        x=1:cy;
```



```

else
    x=1:ry;
end
elseif nargin==2
    [ry,cy]=size(y);
else
    error('One or Two Inputs Are Required.')
end
if ndims(y)~=2
    error('N-dimensional Data is Not Supported.')
end
if min(size(x))>1
    error('X Must be a Vector.')
end
x=x(:); % make x a column
if ry==1 % y is a row vector, make it a column
    yisrow=true;
    y=y.';
    ry=cy;
    cy=1;
else
    yisrow=false;
end
if length(x)~=ry
    error('Length of X Must Match Length of Y or Rows of Matrix Y.')
end
if length(x)<3
    error('At Least 3 Data Points are Required.')
end
dx= repmat(diff(x),1,cy);
dy=diff(y);

dx1=dx(1:end-1,:);
dx2=dx(2:end,:);
dxs=dx1+dx2;
dy1=dy(1:end-1,:);
dy2=dy(2:end,:);

a=(dy2./(dx2.*dxs) - dy1./(dx1.*dxs))/3;
b=(dy2.*dx1./(dx2.*dxs) + dy1.*dx2./(dx1.*dxs))/2;
c=y(2:end-1,:);

i1=(a.*dx1-b).*dx1+c).*dx1; % left half integral
i2=((a.*dx2+b).*dx2+c).*dx2; % right half integral

z=zeros(ry,cy); % pure cumulative Simpson
z(2:2:end-1,:)=i1(1:2:end,:);
z(3:2:end,:)=i2(1:2:end,:);
z(end,:)=i2(end,:);
z=cumsum(z);

% z=[zeros(1,cy);... % original algorithm here has ~2X error
% cumsum([i1(1,:); (i1(2:end,:)+i2(1:end-1,:))/2; i2(end,:)])];

if yisrow
    z=z.';
end
end

```

vectorfix.m

This file reduces the size of the vector containing the numerical solution of the equation of motion.

```
tn=[];  
yn=[];  
cont=1;  
hert=1/f;  
  
while hert >= t(cont) && cont <= 50  
    cont=cont+1;  
    tn=[tn,t(cont)];  
    yn=[yn,y(cont)];  
  
end
```