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UNIVERSITY OF CALGARY

A Study on the Flow Behavior of Microbubbles in Capillary Tubes

by

Mohammad Mehdi Shams

A THESIS

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Abstract

Recently microbubbles have gained an enormous attention in many fields such as drug delivery and ultrasound imaging, oil reservoirs either as sealing agents or means of mobility reduction and CO_2 sequestration. All of these applications involve microbubble flow thorough capillary environments. This study offers a better understanding of microbubbles' flow behavior in capillary tubes to provide a fundamental understanding of their flow in capillary environments.

Stabilized microbubbles have been injected to a capillary tube in which the pressure drop of the flow has been monitored by a differential pressure transducer. To calculate viscosity, theoretical viscosity equations have then been applied to the pressure drop. Besides, the pressure drop was also used to determine wall friction factor. The effect of capillary tube diameter, capillary tube length, quality of microbubbles and flow rate on the viscosity and wall friction factor of the mixture has been assessed.

The data was then used to develop a correlation to regenerate experimental viscosity data of microbubbles, which represents the experimental viscosity data with an absolute average relative deviation (AARD) less than 1.3 %. It was also found that friction factor of microbubble have a linear relationship with non-Newtonian Reynolds number.

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Last but not least I would like to thank Natural Science and Engineering Research Council (NSERC) for their financial support.

Dedication

This thesis is dedicated to my parents.

I would like to dedicate my thesis with all my heart to my mother that has dedicated her life to me and has given me her infinite love.

I would like to dedicate my thesis to my father because of the faith he has in me and the encouragement he has given me throughout my journey in life including my Master's study.

I hope this thesis would appreciate part of what they have done for me. I hope this thesis makes them proud.

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List of Symbols, Abbreviations and Nomenclature

Symbol	Definition
a	Conillant Number Dimensionless
Ca	Capitary Number, Dimensionless
D	Capillary Diameter, <i>cm</i>
K´	Consistency Constant, <i>poise.sⁿ⁻¹</i>
L	Capillary Length, m
m	Constant
m_0	Constant
<i>m</i>	Mass Flow Rate, gr/s
n	Constant, Dimensionless
n´	Flow Index, Dimensionless
r	Capillary Radius, cm
Re	Single Phase Flow Reynolds Number, Dimensionless
Re _c	Continuous Phase Reynolds Number, Dimensionless
Re _n	Non-Newtonian Reynolds Number, Dimensionless
V	Average Fluid Velocity, cm/s
x	Constant, Dimensionless
ΔP	Pressure Drop along the Capillary Tube, <i>dyne/cm</i> ²
$ ho_c$	Continuous Phase Density, g/cm^3
μ_a	Apparent Viscosity, poise
μ_c	Continuous Phase Viscosity, poise
$\mu_r(\mu_{a\prime}\mu_c)$	Relative Viscosity, Dimensionless
ϕ	Bubble Void Fraction, Dimensionless
ϕ_r	Reference Bubble Void Fraction, Dimensionless
σ	Interfacial Tension, dyne/cm
τ	Shear Stress, <i>dyne/cm</i> ²
γ	Shear Rate, s^{-1}
F	Wall Friction Factor, Dimensionless

Chapter One: Introduction

Microbubble is an aggregate of micron sized spherical bubbles suspended in the bulk liquid and stabilized with a capsule of a thin surfactant. This makes microbubble different from conventional foam in structure and in fluid mechanical properties. Foams are essentially gas-liquid-gas system with polyhedral structure wherein thin lamellae of liquid encapsulate the gas phase. Microbubbles' properties result in a system with high gas liquid interfacial area which will flow readily.

The concept of microbubble was first introduced in 1971 [1]. Microbubbles' characteristics and properties were described in detail and the possible applications were predicted in a book entitled Foams and Biliquid Foams-Aphrons written by Sebba [2]. He described microbubbles as bubbles approximately 10-100µm in diameter. A microbubble is made up of a spherical core. This core is encapsulated in a thin aqueous shell. The thin liquid shell (film) is consisted of two surfaces separated by a short distance. The colloidal properties of microbubbles are due to the presence of surfactant molecules at the thin film [2].



Fig. 1 Structure of microbubbles [2]

Microbubbles differ from conventional foams. The shell of a microbubble consists of a tri-layer of surfactants. However, a conventional bubble is made of a surfactant monolayer. As shown in Fig. 1 the air core is supported and separated from the viscous layer by the inner layer of surfactant molecules. On the other hand, the viscous layer is supported with the outer layer which is hydrophobic outwards and hydrophilic inwards. It is also believed that there is another layer of surfactant molecules hydrophobic inwards and hydrophilic outwards for the bubble to be in contact with the bulk water. A surfactant stabilized conventional bubble is simply separated from its aqueous phase by a thin surfactant film. Thus, a microbubble (

Fig. 1) has a hydrophobic outer boundary while a conventional bubble has a water wet or hydrophilic boundary [2].

As the main component of microbubbles, a surfactant is used for three main reasons. Firstly to produce the surface tension to contain the microbubble as it is formed. Secondly to build the multilayer wall which will stabilize the microbubbles. Surfactants also create interfacial tension to form a non-bonding network capable of bridging openings. For these purposes both ionic (cationic or anionic) and nonionic surfactant can be used [2-4]. However, it is suggested that ionic surfactants will generate more stable microbubbles compared to non-ionic surfactants [5]. Some other investigators have suggested that stability of microbubbles increases with the length of the alkyl chain in ionic surfactants [6]. In general it has been suggested that non-ionic surfactants generate bubbles with smaller diameter than those generated by the ionic surfactants [7].

Wan et al. investigated microbubbles produced by water-soluble surfactants [8]. They found out that bubbles produced with a single or a mixture of water-soluble surfactants, SDS, SDeS, RE-960,ES270, D8390, Tween 80, and Triton X100, had short life spans (less than an hour) and

large sizes (30-100 µm). These bubbles formed foam rapidly when left standing. They also conducted experiments on hydrophobic surfactants. These surfactants (Span 60 and Span40) yielded no bubbles. They concluded that stable microbubbles are generated using a combination of water-soluble surfactants and solid hydrophobic surfactants. Wheatley et. al. [9] have also reported stable microbubble generation using a combination of solid hydrophobic surfactants Span 60 and 40 and water-soluble Tween surfactants (Tween 20, 40, 60, 65, and 80). They concluded that liquid hydrophobic surfactants especially undecanol and dodecanol do not form stable microbubbles. All the previous results indicate that a solid hydrophobic surfactant in combination forms a solid-condensed layer at the gas water interface. Diffusion of gas can be slowed by the tight packed layer. Moreover, the surface tension of the mixed tri-layer has been reported to approach zero [10].

Stable microbubbles have been found to have various applications such as biological purposes, water purification, sealing agents in underground geological formations and means of enhanced oil recovery. Most of the applications proposed for microbubbles consist of their usage in capillary environments either in porous media or in human body. Therefore, flow behavior of microbubbles in capillary environments plays an important role in their utilization.

The overarching objective of this research is to:

- evaluate the flow behavior of microbubbles by investigating their viscosity
- assess the friction factor of microbubbles in capillary tubes as the important parameters in microbubbles' behavior.

Chapter Two: Literature Review

2.1 Microbubble Application

The special structure and stability of microbubbles have made them suitable for many applications. Their small size increases the active surface area required to adsorb contaminants in microflotation purposes [11]. Stability under pressure and the ability to act as contrast agents against ultrasound waves have made microbubbles an appropriate candidate for drug delivery and in vivo ultrasonic imaging [12]. Their stability under pressure also results in a sealing ability, which is utilized in drilling muds to reduce mud loss in high permeability reservoirs [13]. Beside their stability, the increase in viscosity due to their presence in aqueous solutions can result in an increased sweep efficiency, which has been exploited in waterflooding enhanced oil recovery (EOR) as a replacement of polymer injection [14]. Another possible application is CO₂ sequestration for greenhouse gas reduction by underground injection. In this type of injection, the stability and small size of microbubbles are used to provide a leak-free injection with higher dissolution in saline aquifers [15, 16]. It has also been reported that, in some reservoirs, the gas derived from oil due to a decrease in pressure, will form microbubbles instead of conventional large bubbles. These microbubbles have been found to preserve the pressure inside of the reservoir and, thus, increase oil recovery. This form of microbubbles in oil is called foamy oil [17]. Each application mentioned above is based on a special property of microbubbles. More information about each application and the property that is applied in that application is explained in the following pages.

2.1.1 Ultrasound Contrast Agent

Gramiak and Shah [18] were the first to suggest bubbles in 1968 to be used as ultrasound contrast agents (UCAs), to increase the quality of sonograms in medical imaging (Fig. 2)[19-21]. The quality of ultrasound imaging by microbubbles depend on compressibility and diffusivity of

gas to bubbles [22], viscosity and density of the surrounding medium, the frequency and power of ultrasound, and the size of the bubbles [23-25]. To avoid safety problems microbubbles must be smaller than about 10 μ m to ensure smooth passage through blood capillaries [27-29]. The most suitable size range of microbubbles is 3-5 μ m [30].

Microbubbles can also be burst by ultrasound waves to combine imaging and therapy (Fig. 3). This will lead into image-guided drug delivery [31-33]. It has been mentioned by Mitragotri [34] that a broad range of ultrasound frequencies (20 kHz-1 MHz) are used in medical applications.

The most important parameters for microbubbles' acoustic response are their size and shell features. Size and shell thickness effect scattering intensity, absorption of acoustic energy and destruction threshold. Shell damping effect is directly related to resonance frequency of polymer shelled microbubbles which decreases with an increase in bubble size [35]. Microbubbles compared to free gas bubbles have an increase in resonance frequency due to their stiffness and an increase in damping due to its viscosity.



Fig. 2 Sample of microbubbles applied to ultrasound imaging [36]



Fig. 3 Bursting process of microbubbles in drug delivery [37]

Sonic cracking is the major mechanism for bubble destruction in microbubbles with a rigid thin shell [38, 39]. Sonic cracking is the ultrasound-induced formation of shell causing gas to escape from the microbubbles. Thus, understanding the interaction between microbubbles and ultrasound at difference frequencies is important.

2.1.2 Microflotation

Microflotation is known as a removing process for various colloidal pollutants from water and water wastes. This method was first introduced by Rubin and Cassell [40]. The advantages of this process against other flotation processes are requiring very low gas flow rates, extremely small bubbles, and heterocoagulation of the colloidal dispersions with a hydrous metal oxide. The last property is strongly pH dependent. Floated material can be collected by addition of a collector, such as a long-chain fatty acid or amine. It will produce a stable surface phase and a thin layer of relatively dry foam. In order to have smaller (~50 μm diameter) and more stable bubbles, frothing agents (i.e. alcohols) are added. Since bubbles are smaller in microflotation and flow rates are lower, a larger gas/liquid interfacial area per unit of gas flow rate is produced with no severe agitation.

In contrast most of the conventional foam separation techniques require higher gas flow rate to have sufficient interfacial area for adhesion of solid particles to bubbles. This will cause turbulence at the foam/liquid interface. To prevent floated particles from redispersion, a tall foam column is also required.

The early work regarding microflotation was used for the removal of bacteria and algae from water [41, 41]. More studies showed that microflotation could be used to remove B. Cereus [42], colloidal constituents of tea [43], humic acid [11], colloidal silica [44], illite [45], titanium dioxide [46], and polystyrene latex [11] efficiently. Microflotation is a nonselective method capable of colloidal dispersed material removal commonly found in natural and waste waters

[47]. This method is also applicable efficiently over a broad pH range which depends on the choice of the coagulant [48].

It is well known that the most important parameter in determining efficiency of a flotation or foam separation process is bubble size.

2.1.3 Drilling Mud

Conventional drilling fluids very often generate circulation loss and differential sticking problems when used to drill the depleted or low pressure reservoirs. Extra equipment and special protection measures are required to use aerated drilling fluids or underbalanced drilling. Besides, it may be unsuccessful to use these techniques to provide the hydrostatic pressure necessary to stabilize normal or high pressured formation above the low reservoir pressure. Microbubbles have been developed to solve loss circulation, borehole stability and differential sticking problems in low reservoir pressure. In these fluids conventional bridging agents to seal the loss zone have not been used. These drilling fluids combine certain surfactants to create microbubbles. These bubbles compared to conventional air bubbles have advantages in aerated mud system. The air is purposely at a very low concentration incorporated into the bulk fluid. The microbubbles are entrained by conventional mud-mixing equipment up to level dictated by the concentration of microbubble generating surfactants [49]. The entrained air is converted to stable microbubble by surfactants. Stability of microbubbles at downhole conditions will set up inner bridging in low pressure formations. This will prevent uncontrollable fluid loss and prevents formation damage.

Microbubbles were first introduced to petroleum drilling industry by Brookey [50]. The first applications of microbubble were in West Texas for a horizontal re-entry well drilled through fractured dolomite in the Fusselman field. After that, microbubbles have been used in drilling thousands of depleted reservoirs without any loss circulation problem [49-58].

The typical microbubbles used in drilling muds are from 10 μm to 100 μm in diameter. These bubbles can survive recirculation through mud cleaning system. Most microbubbles are not removed even by fine screen shale shakers or flow line cleaners. Moreover, their little mass, will make them retained even in hydrocyclones or high-speed centrifuges. Microbubbles also do not interfere with downhole tools such as mud motors which will make them ideal for directional and horizontal applications. In microbubbles, due to little amount of air entrained in the base fluid (15%) their density is similar to density of the base fluid. High values of viscosity in lowshear-rate helps to set up bridging and sealing formation [50, 53].

Stability of microbubbles depends on thickness and viscosity of the encapsulating shell. The shell must have a certain minimum thickness. Thickness is important for microbubbles in circulation where their volume will change with pressure change according to Boyle's Law. The shell will break if it becomes excessively thin. This may happen on expansion when exposed to a very large pressure drop [2, 59]. The water/film shell is not stable if it is thinner than four microns or thicker than 10 microns [51]. Apart from thickness, shell viscosity is a very important criterion for microbubble stability. To prevent Marangoni effect that causes diffusion of water out of the shell into the bulk liquid, the shell must have a minimum viscosity. This thins and destabilizes the shell [2]. The rate of water transferred is proportional to shell viscosity. Therefore, it is required to add a viscosifier such as biopolymer. The viscosifier is also effective to slow the flow into loss zones [49, 60].

The most common type of microbubble used in the field is a polymeric water-based system. There has also been an alternative water-based clay and a non-aqueous-based microbubble drilling fluid (ester-based microbubbles) developed [57]. The difference between the two systems is the continuous phase which is water (fresh water or brine) in water-based microbubble system, and oil or synthetic fluid in oil-based microbubble system [51, 59-61].

Another advantage of microbubbles over conventional foams in drilling muds is the elimination of corrosion due to presence of air. When drilling with air systems and aerated fluids, corrosion becomes a major problem. Traditionally to prevent this problem from happening, attempts have been made to minimize air entrainment. The air in microbubbles is incorporated into the bulk fluid at a very low concentration. The surfactants in the fluid convert the entrained air into microbubbles. The oxygen from the air in the core is lost via chemical reaction with various components in the fluid. This process usually takes minutes and result in the microbubbles being filled mostly with nitrogen. Thus, corrosion caused by microbubbles is negligible. This has been proven in the field where with the lack of a corrosion inhibitor, corrosion rates were very low in wells even with the presence of high concentrations of H_2S [52].

The effects of microbubbles on the flow through permeable and fractured media have been determined by various laboratory techniques. When the drilling fluid enters a formation, the microbubbles expand to a small extent. More importantly, they will move forward rapidly to concentrate at the fluid front. This will create a "microenvironment" that separates the borehole from the formation pressure.

When a hydrophobic microbubble wants to enter a water wet interconnected microfracture/pore network in permeable formations, capillary pressure resists its invasion [49, 51, 54]. The resistance to flow of microbubbles and the carrier fluid into formation openings is dependent on the size of the openings and the degree of hydrophobicity of the microbubble outer shell. Small openings and strongly hydrophobic/hydrophilic microbubbles promote sealing. On the other

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hand, very large openings or fractures will have little or no capillary pressure. Therefore, no sealing may be possible except at the fracture tip [51, 60].

Microbubbles are brought together at the openings of low-pressure zones when the drilling bit exposes a depleted formation. There, a portion of the energy stored within each microbubble is released, making it to expand until the internal and external pressures on the wall of the microbubble are in balance [55, 60, 62, 63].



Fig. 4 Microbubble's aggregation at formation's opening [64]

As the microbubbles get crowded in the formation's opening, external aggregation will happen and the viscosity will increase (Fig. 4). A solid free bridge will be formed by the microenvironment created by this phenomenon. Another benefit of the non-conventional internal seal is its effect on differential sticking. This will exhibit a gradual pressure drop from the annulus to the seal interface with the reservoir fluids. This translates into a considerable reduction in risk when employing costly downhole tools during well construction in high-annular and low reservoir-pressure applications [55, 60]. Microbubbles will migrate with the pressure gradient from the wellbore to the formation when wellbore pressures exceed formation pressures. The microbubble will again move with the pressure gradient, from the formation into the wellbore if wellbore pressure is lowered to below the formation pressure. Since surfactant is consumed on the drill cuttings and/or in the borehole it is important to consider the depletion. Inadequate surfactant concentration can lead to increased downhole losses.

The microbubble drilling fluid technology has been successfully applied in drilling vertical, horizontal and inclined well. They have been applied to operations in South America, North America, Africa, Far East, Eastern Mexico, Venezuela, North Sea, North Texas, and West Texas [49-51, 53, 55-58]. Microbubbles have been used in hundreds of wells worldwide successfully in depleted reservoirs in mature oil and gas fields, high-permeability formations and micro fractured rocks [51, 61].

2.1.4 Enhanced Oil Recovery

Current chemical flooding technologies are dealing with inadequate sweep efficiencies and unfavorable mobility ratios. This will leave much of the recoverable oil left untouched in the pores of the reservoir. Numerous methods have been proposed to solve the low sweep efficiency. For instance, different formulations of polymer and surfactant base fluids have been used to solve this issue and there have been varying degree of success. The idea of using microbubbles for enhanced oil recovery were suggested when the sealing ability of microbubbles was proved in drilling muds.

In drilling muds when the fluid enters the formation, the microbubbles will concentrate at the fluid front with rapid forward movement of microbubbles. This phenomenon leads to a microenvironment that isolates the bulk fluid from the formation. This inhibits invasion of fluid into formation. Islam and Chakma [65] investigated the role of the microbubbles as a displacing agent in the heavy oil recovery process. By injecting externally generated microbubbles in a straight capillary tube and injecting a free gas in a porous medium, they found that the microbubbles increased recovery factor substantially. Bjorndalen et. al. [66] observed that resistance of microbubbles increased continuously as more microbubbles are injected to porous media. This proves microbubbles effective blocking property. This flow characteristic of microbubbles is the main motivation of using microbubbles for EOR applications. Samuel et. al. [67] have investigated the capability of microbubbles in enhanced oil recovery. They find out that the total recovery achieved by microbubbles is lower than conventional polymer injection. However, microbubbles require less pressure to be injected than polymers. The breakthrough time of microbubbles is also higher than polymers with more oil recovered by microbubbles at the time of breakthrough. This illustrates improved sweep efficiency of microbubbles compared to polymers (Fig. 5).

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Water Followed by Polymer

Fig. 5 Comparison of polymer flooding and microbubble flooding [67]

2.1.5 Foamy Oil

Smith [68] has suggested that micro bubbles have a significant role in solution–gas drive in heavy oil. These microbubbles are called foamy oil. On the other hand, other investigators believe this phenomenon is of less importance [69-71]. Quantitative investigations on the role of micro-bubble are rare [72]. The main property of microbubbles is the small size of bubbles that are smaller than the average pore size and can flow with the oil. Generation of microbubbles in the porous media is reported by Ward et. al. [73]. Smith [68] was the first to describe performance of heavy oil reservoirs under favorable solution-gas drive. He described the flow in such solution–gas drive systems in viscous oil reservoirs to have tiny gas bubbles moving with the oil. Once microbubbles are formed neither stay behind nor rush ahead but move with the oil. Claridge and Prats [74] found out that if gas bubbles are formed below the bubble point pressure asphaltene particles separated from the oil will stabilize them. They hypothesized this separation to result in the remainder of oil with a significantly lower viscosity. The lower viscosity will make this oil to flow with the stabilized microbubbles. At pressures immediately below the bubble point pressure, Treinen et al. [75] observed a reduction in produced GOR. It was concluded that reduction is due to the trapped gas bubbles which do not flow with the oil. Pooladi-Darvish and Firoozabadi [69] indicated in their heavy and light oil depletion experiments that the pressure gradient across the porous medium increased as the average pressure was reduced to below bubble point pressure. They suggested that mobility of the oil is reduced due to the formation of the gas phase. At higher gas saturations gas flow was observed. Their results confirmed previous understandings of traditional solution-gas drive. In this point of view gas bubbles are formed and trapped in a porous media until bulk gas flow is formed at a critical gas saturation.

Bora et al. [76] visually examined solution–gas drive in heavy oil at a pore scale. The main objective of the examination was to see if large populations of micro bubbles happen in the oil and gas flow mechanism. They used crude oil with and without asphaltenes. The tests could not confirm the existence of stable micro bubbles. Therefore, asphaltenes were not confirmed as stabilizers of microbubbles. However, Bora et. al. [76] has shown that compressibility of the oil before the appearance of the first visible bubbles is significantly greater than that of the live oil but smaller than the gas compressibility during the early stages of the depletion and before the appearance of the first visible bubbles larger than 2 μm has enhanced the production. Maini [70] related high recovery factor observed in the fast experiments to the large pressure gradients instead of the idea of the explosive nucleation of a large number of bubbles. Shahabi et al. [77] conducted a series of depressurization experiments, with and without pre-existing micro bubbles, using a glass micro-model. He did not observe foamy oil behavior.

Opinions about microbubbles and their importance differ among researchers. This could be related to various experimental conditions in different studies.

2.1.6 Carbon Dioxide (CO₂) Sequestration

Mismatching between sources and sinks is the most serious problem of the worldwide deployment of carbon capture and sequestration (CCS) to prevent the global warming. Greenhouse gas and supercritical CO₂ fluid can only be stored in geological formations that contain impermeable caprocks (clay, shale, etc.) and porous reservoir rocks (sandstone, conglomerate, etc.). Geological structures that can store large mass of buoyant CO₂ are often far from greenhouse gas emitters such as fossil-fueled power plants. Novel method of carbon microbubbles sequestration technology [78] is introduced to inject large amount of CO₂ safely

and economically into ordinary underground rocks. In this method there is no need for the special geological structures that can trap large buoyant bodies of the greenhouse gas.

Injection of large amount of CO_2 into underground formations will result in a big underground CO_2 bubble. This bubble is a region of connected pore occupied by a fluid mostly consistent of CO_2 . Dependent on the pressure and temperature of the reservoir the pure CO_2 is gas or liquid or supercritical fluid. Since at the ordinary underground pressure and temperature condition the CO_2 fluid (gas or liquid or supercritical fluid) is lighter than the groundwater, it will gain the big buoyant force. At Sleipner, Norway in the first large-scale CO_2 sequestration project uprising of large CO_2 bubble was observed in the highly permeable sandy aquifer [78]. For safe storage of large CO_2 fluid the perfect cover of underground geological structure such as a dome of caprock is necessary. Natural gas and oil deposits have long term geological traps that have contained the buoyant fluids longer than several million years. In the Sleipner aquifer it was also suggested that undetected small gaps of shale layers may have provided pathways of CO_2 [79]. For the capsrock that have not been proven naturally, there is no effective technology that could check out their completeness.

The difference between the inside and outside of CO_2 bubbles will cause a large excess fluid pressure at the top. The excess pressure in the CO_2 reservoir cannot exceed the interfacial threshold pressure for the CO_2 breakthrough of caprocks. If the excess pressure due to buoyancy reduces the effective stress enough to cause the shear instability of underground rocks the fluid fracturing of caprocks and fluid-induced earthquakes may occur at the top of large CO_2 bubble. However, the fluid fracturing and induced earthquakes are unlikely to happen around microbubbles. This is because only a very small excess fluid pressure builds up at the top of microbubbles.

Implementing CO₂ microbubbles can drastically reduce the size of CO₂ bubbles. This injection technology generates numerous uniform tiny bubbles of carbon dioxide and/or other greenhouse gases smaller than ten micrometers in diameter in water. The mixture of the CO₂ microbubbles and water is injected through the wall of deep borehole into tiny pore spaces of underground rocks. The microbubbles less than several 10 micrometers in diameter tend to shrink and quickly solve into water. The greenhouse gas microbubbles do not tend to join together to form large bubbles that have large buoyant force in groundwater. In the aquifer rocks, CO₂ microbubbles percolate deep into tiny pores and cracks [80]. Many carbon microbubbles are trapped in pores of rocks with interfacial force and capillary effect. At the underground high pressure, CO₂ microbubbles dissolve into the groundwater rapidly. A water-saving version of carbon microbubbles injection technology is the underground injection of dispersed tiny droplets of CO₂ liquid [81]. CO₂ solution tends to flow downward, because the CO₂ solution is heavier than the primary groundwater. Carbon dioxide gets dispersed into underground rocks as microbubbles prevent the large carbon dioxides bubbles (gas or supercritical fluid) to rise in partially confined aquifers and even in unconfined aquifers. Residual gas (interfacial and capillary effects) trapping, dissolution trapping, and ionization trapping of CO₂ accelerate with carbon microbubbles injection. Long-term CO₂ storage in solution has been proven in young semi-open unfolded sedimentary basins of natural methane solution deposits (in Chiba, Niigata and Miyazaki prefectures) and natural CO₂ solution deposits (at Isobe and Izumi districts) in unconfined saline aquifers in Japan [82]. Storing CO₂ in saline aquifer storages is the economically prospective option of geological carbon sequestration. This has the huge storage potential of three trillion tons of CO₂ in solution in the worldwide sedimentary basins [83]. For

many urban areas and industrial areas carbon microbubbles sequestration (CMS) in saline aquifers provides the economic option of carbon capture and storage (CCS).

The carbon microbubbles injection can be applied to both small-scale sequestration of greenhouse gases and large scale CO_2 storage. In the coming hydrogen society a large number of relatively small-scale sources of greenhouse gases would be dominant. CO_2 is harmless at low concentrations but it causes the global greenhouse effect. Small-scale carbon microbubble injection provides an essentially safe strategy of greenhouse gas sequestration.

 CO_2 has much higher solubility than nitrogen and methane at high pressure. Mixed gas microbubble injection, such as flue gas, may result in selective dissolution of CO_2 . The carbon capture by microbubbles, may be realized as an efficient and economic option for small-scale CCS without the use of any solvent. In order to have an efficient separation of CO_2 , depth of microbubbles generation in the injection well is important.

Higher permeability is expected from rocks than pores of rocks. However, it becomes extremely low for larger bubbles than pores of rocks for smaller microbubbles. Suitable microbubbles should be smaller than pores of reservoir rocks but larger than pores of caprocks. In the oil shale and tight gas reservoirs, extremely small microbubble (nanobubbles) could penetrate into extremely small pores.

The underground carbon microbubbles injection will result in gas trapping, dissolution trapping, ionization trapping, mineral trapping, microbial trapping and methanogenesis which accelerates the geological sequestration mechanisms by dispersion and quick dissolution into groundwater [84]. Very weak buoyant force of greenhouse gas microbubbles, together with heavy CO_2 dissolution and various trapping mechanisms will make the carbon microbubbles stable in various geological conditions even with some structural imperfection. For instance, depleted

oil/gas reservoirs with abandoned wells, barren geological domes with unproven caprocks, faulted anticlines of sedimentary formations, saline aquifers with incomplete caprocks, large horizontal or monoclinal formations, synclinal sedimentary basins, fractured basalt layers, fractured serpentine bodies, fractured igneous bodies can be used for CO_2 microbubble sequestration.

The quick dissolution of CO_2 microbubbles around the injection well forms the region of acidic interstitial water in the underground reservoir rocks. The porosity of rocks will increase due to dissolution of calcite and decomposition of feldspars and other minerals in interstices of grains by CO_2 -rich acidic water. However, pH of water is increased and CO_2 -water-rock interaction neutralizes as the CO_2 dissolution proceeds. Carbonates in cation-rich and CO_2 -rich water precipitates at high pH conditions and seal the interstices of rocks autogenously. This process happens especially in Mg and Fe-rich mafic rocks such as basalt, peridotite and serpentine. These CO_2 capsules can be formed in large basaltic sheets, ophiolite complex and oceanic crust. Autogenously sealed " CO_2 capsules" can store greenhouse gases permanently.

 CO_2 storage in deep subsea formations has been proposed by Koide et al. [84] indicating the virtually perfect leak-free condition due to the hydrate trapping. CO_2 sequestration in sedimentcovered basalt aquifers on the Juan de Fuca plate have been suggested by Goldberg et al. [85] to securely accommodate more than a century of future U.S. emissions. The autogenous sealing by carbonate trapping and by hydrate trapping provides a promising leak-free storage for vast amount of anthropogenic CO_2 in the oceanic crust.

2.2 Microbubble Generation

Microbubbles of uniform sizes and varying mean diameters can be prepared with techniques such as microfluidics [86] or electrohydrodynamic atomization [87]. On the other hand, microbubbles obtained using high speed mixer [88] and sonication method [89] are prepared easier in bulk scale but have less monodispersity. Various technologies used for synthesizing microbubbles have been reviewed by Stride and Edirisinghe [90]. It is worth noting that using complex processing equipment to create bubbles of uniform size and varying mean diameters does not solve the post-preparation problems associated with the dissolution or growth of microbubbles. Coated microbubbles dissolve and shrink due to the existence of Laplace overpressure. In addition, microbubbles grow and lose their monodispersity in result of coalescence and the Ostwald ripening. Recently, microfluidic devices have been used to create monodisperse size distribution of lipid-protein encapsulated microbubbles with stability of few hours [91]. Lipid-protein [92] and protein-polysaccharide shelled microbubbles can be produced by by microfluidics with a narrow size distribution and a longer term. Size distribution of microbubbles synthesized by sonication, microfluidics, and coaxial electrohydrodynamic atomization techniques have been compared by Stride and Edirisinghe [93]. Zhou et al. have concluded that microbubbles produced by sonication and coaxial electrohydrodynamic atomization techniques have the size range required for intravenous administration [89].

2.2.1 High Speed Mixer

Sebba proposed a generator comprised of a thin solid metal disc approximately 2-3 *cm* diameter, mounted horizontally about 2-3 *cm* below the surface of the surfactant solution [88]. The proposed generator can be seen in the Fig. 6. He described the dimensions not to be critical. The disc was mounted to a vertical shaft connected to an electric motor capable of 7000 r.p.m.. Two rigid baffles made of perspex were also mounted vertically, shaped to encompass the disc and to

give a clearance of about 1cm from the shelf. There were two applications for the baffles. When the disc spins above about 4000 r.p.m., the hydrodynamic forces produced by the disc will create strong waves on the surface. The waves strike the baffles, and will form small bubbles when they re-enter the water. Secondly, the baffles will reduce the tendency to form a vortex. The gas entrained in a vortex will form bubbles with a large size distribution. However, it is desirable for microbubbles to be as uniform as possible. There is a critical speed of revolution which below that, microbubbles are not produced. When that is exceeded, the microbubbles are generated remarkably fast.



Fig. 6 Proposed microbubble generator by sebba [88]
For instance, within a few seconds one litre of surfactant solution in a three liter beaker will be converted into microbubble suspension containing over 60 percent of gas. When the microbubbles have been produced, the spinning speed can be reduced just to keep the bubbles moving. This can make the microbubbles maintained as long as one hour without much deterioration. The microbubbles are about 25 μm in diameter and because of the encapsulating surfactant film as long as they are completely immersed in the water they will not coalesce. If the microbubbles start to rise to the surface where they form conventional foam, it is needed is to increase the speed of the spinning disc to above the critical speed. Within a few seconds the foam will be destroyed and converted into microbubbles.

Vibration is a problem with such a rapidly revolving system. Therefore, it is necessary to clamp the shaft in a bearing housing to hold the shaft in a framework, otherwise there may be creep. If it is desired to generate the microbubbles in larger capacity all that it is only required to surround the disc and baffles by a hollow tube leaving about 3 *cm* clearance with the baffles. This will keep the surface wave still produced and the spinning disc will drive the microbubble generated under the lower end of the tube to mix with the remaining solution.

2.2.2 Ultrasound Cavitation

Most lipid or protein encapsulated microbubbles are prepared with sonication that result in the formation of polydispersed microbubbles. Polidispersity will result in reduced efficiency of microbubbles' imaging and therapy. Clinical imaging systems aim a small percentage of a microbubble population and operate at a limited frequency bandwidth. On the other hand, in molecular imaging and targeted drug delivery monodispersed microbubbles will elucidate quantifiable information. Lower dose of microbubbles are required to be injected into patients with better drug delivery. Therefore, it is important for the microbubbles to have a narrow size distribution for effective drug delivery and contrast imaging.

Ultrasonic irradiation of liquids can produce cavitation. This will cause the formation, growth, and implosive collapse of bubbles [94]. Transient hot spots with enormous peak temperatures are created by the compression of such bubbles [94, 95]. It might be thought that this localized heating which causes thermal denaturation of the protein is responsible for microbubble formation. In this case, microbubble yields for Ar versus O_2 or N_2 should be more due to the effects of dissolved gases on cavitation (because of decreased heat capacity ratios, C_p/C_v). However, it is equal for the latter two gases. Experimentally, microbubbles are synthesized in high concentrations only when the reaction is run under O_2 or air. Sparging Ar or N_2 into solution before ultrasonic irradiation diminishes the formation of microbubbles substantially. Thus, proteinaceous microbubbles are resulted from specific chemical reactions involving O_2 not from heat denaturation of the protein.

A novel post-synthetic sonication approach to control the size distribution of ultrasonically synthesized microbubbles has been introduced by Zhou et. al. [89]. They have shown that the size distribution of microbubbles can be selectively modified by post-sonicating an aqueous dispersion of the microbubbles at various frequencies.

2.2.3 Microfluidic T-junction

In microfluidic devices it is possible to have control on small volumes of fluids and understanding of multiphase flows. There have been numerous methods to generate Liquid-liquid dispersions in microfluidic devices. These methods include geometry dominated breakup [96], crossflow rupturing through microchannel arrays [97], hydrodynamic flow focusing through a small orifice [98, 99], and two-phase crossflowing rupture streams in T-junction microchannels [100-105]. These investigators found that parameters relevant to the drag force of the continuous phase and the momentum of the dispersed phase, such as flow rates, viscosities, and the interfacial tension affect droplet formation of the dispersed phase.

However, fewer studies have dealt with gas–liquid dispersions. Garstecki et. al. [106, 107] described the capabilities of a flow-focusing device incorporated directly into a microfluidic chip that delivers monodisperse bubbles at frequencies exceeding 10^5 bubbles per second. In their work, the bubbles were highly uniform. A method of geometry dominated breakup in microchannel (MCs) plate has been reported by Yasuno et. al. [108]. Xu et. al. described microbubble formation to be based on the spontaneous transformation caused by surface tension [109]. They described the bubble size to be affected by viscosity, flowrates (Fig. 7) of gas and liquid, and the dispersing agents. They also found bubble size to be independent of the interfacial tension.



Fig. 7 Effect of liquid flow rate in constant gas flow rate on bubble size in microfluidic T-junction method a) Air-phase flow rate= 25µL/min, aqueous phase flow rate= 80µL/min; b) Air-phase flow rate= 25µL/min; aqueous phase flow rate=180µL/min

[109]

2.2.4 Coaxial Electrohydrodynamic Atomization

Microbubbles are known for medical application (drug delivery, ultrasound imaging). For These applications, a well-defined microbubble size distribution is required. In drug delivery, in order to ensure correct dosage, drug content and destruction threshold are critical. Common microbubble preparation methods such as agitation by high speed mixer and ultrasound cavitation do not provide the required level of size distribution control. Microbubble production via agitation or sonication, results in a broad size distribution. It is necessary to filter out microbubbles having diameters >10 μm to prevent the risk of causing an embolism in vivo. The remaining size distribution is still relatively broad after filtering. This will result in a wide range of microbubble resonance frequencies. Also individual microbubble coatings' have a wide variation in their properties. This will change their dynamic and acoustic response significantly [110]. For example, stiffness of the coating has an effect on microbubble cross-section scattering. This will make it hard to predict and control the microbubbles' response. In particular it makes it extremely difficult to predict the point at which the coating will rupture.

In Electrohydrodynamic atomization (EHDA) liquid or a suspension is passed through a capillary needle at a controlled flow rate with electric field applied to it to undergo atomization [111]. Atomization is influenced by the liquid properties such as surface tension, viscosity, electrical conductivity, density, relative permittivity and process parameters such as flow rate and applied voltage. Droplet size decreases with the applied voltage and increases with the flow rate. It is necessary to select the right values for these two parameters to achieve the minimum size of a droplet [112]. Edirisinghe et al. were first to use electrostatic atomization of suspensions. It has now been adopted for the spray deposition of biomaterials [113], forming of ceramic foams [114], direct writing [115] and for many other industrial applications.

Farook et al. proposed a method to overcome the disadvantages of the existing methods by using electrohydrodynamics [116]. Their method offers significant benefits where size and uniformity is a key property. In other words, this method facilitates microbubbles as advanced diagnostic and therapeutic agents.

Co-axial electrohydrodynamic atomization (CEHDA) is a developed method of EHDA. This method is an industrially viable microbubbling technique proposed by Farook et al. [116]. A pair of co-axial needles is used in CEHDA, in contrast to EHDA, which uses a single needle (Fig. 8). Furthermore, two different liquids or suspensions are pumped through two concentrically placed needles in CEHDA. Liquids are capable of experiencing various modes of atomization subject to an applied voltage and the co-flowing liquids. Near-monodispersed compound aerosols are formed as a result to the eventual jet break-up with the outer material surrounding (Fig. 9). Encapsulation of food additives, targeted drug delivery and material processing are most important applications of this phenomenon [117]. Moreover, encapsulation of suspensions of zirconia by suspensions of alumina has been utilized to prepare composite ceramics and bio-microstructures and vice versa [118].



Fig. 8 Equipment used for co-axial electrohydrodynamic atomization [116].



Fig. 9 Microbubble formation by co-axial electrohydrodynamic atomization [116]

2.3 Previous Studies on Rheology and Friction Factor of Foams and Bubbles

2.3.1 Viscosity and Friction Factor of Foams in Porous Media and Tubes

Foams are characterized by the large volume fraction (generally greater than 74% in a foam of uniform cells) of the non-continuous fluid component. The word foam is used in a broad sense in much of the literature. Foamlike dispersion is more descriptive of this composite substance and has been suggested by Heller et. al. [119]. Sebba [88] has suggested the use of the phrase biliquid foam. Due to the crowded nature of the discontinuous phase, cells of foams are incapable of significant independent motion. However, foams can flow.

Most of general literatures on foam have emphasized on the structure and the surface chemistry of films that separate the cells of internal-phase fluid. In some of the porous media work, various parameters affecting the mechanism of foam flow is emphasized, including the type and concentration of surfactant in the aqueous phase, the volume fraction of the foam quality, and the texture (average bubble size). Some other investigations regarding foam flow in porous media have been done [120-128]. However, no general agreement has been reached.

The result of most of the works done on measurement and analysis of foams have been put in terms of the traditional rheological parameters developed for use with other fluids. For instance, effective or apparent viscosity has been used to describe the frictional forces retarding the flow of foam. Coefficients are computed according to the particular geometries in which they were derived; therefore, they are often not appropriate at a different scale. Unlike those measured in simpler fluids there is experimental evidence in these papers that the apparent viscosity values for foams, are geometry dependent. However, it has been indicated that the relation between wall friction factor and Reynolds number in foams is identical to single phase flow [129].

Factors important in the design and interpretation of foam flow experiments that influence the flow behavior of foams are as follows: (a) the ratio of (mean bubble size)/(flow channel size); (b)

size distribution of bubbles; (c) flow-induced anisotropy of bubble distribution; (d) foamantchannel wall interactions (slip, adsorption); (e) characteristics of flow geometry and flow rate; (f) quality (or nonpolar-phase volume fraction) of foam; (g) properties of the two fluid phases; (h) absolute pressure, (this effect generally arises from a difference in compressibility of the two phases); (i) physicochemical nature and concentration of foamant; (j) interfacial rheological properties of foam lamellae and their variation with (aging) time (these would affect the stability of foam).

2.3.1.1 Viscosity of Foams in Bulk

Rheological properties of foams have been investigated in a number of rotational viscometric devices, such as the Brookfield viscometer [129], the Fann VG meter, and the Epprecht coaxial-cylinder viscometer [130] and modified versions of a Fann coaxial-cylinder viscometer [131, 123] and of a cone-and-plate viscometer [132]. In order to measure limiting values of viscosity at high rotational speeds Sibree used a specially designed concentric cylinder viscometer [133].

A major experimental problem perceived when using these viscometers is that samples are subjected to shearing for a finite period of time until steady-state measurements of torque can be made. Unfortunately, foam bubbles in contact with the rotating solid surface may collapse or have rearrangement in their network. This will cause a partial discontinuity in the transmittal of angular motion; thus, the torque measured is not indicating the foam sample undergoing shear. This is similar to the slippage phenomenon in certain non-Newtonian fluids at the solid-liquid interface. Besides, during the course of testing, drainage of liquid may occur which will change the quality of the foam being sheared [131], unless very dry foams (quality exceeding 85% or 90%) are examined. Since the "time of shearing" has a pronounced influence on torque readings obtained with wet foams, it has been difficult to achieve reproducibility of data in practice.

To minimize drainage a continuous flow of foam was provided to the viscometer [123]. The stator and rotor surfaces were modified from the usual smooth cylindrical shape by installing thin, long fins to minimize slippage. The interpretation of the data in this case has not been straightforward either. For instance, measurements of apparent viscosity using this modified Fann VG meter were found to lie between 50 and 500 cp for aqueous foams having gas volume fractions in the range 0.70-0.96. These values are in contrast with order-of-magnitude compared to lower values (3-8 cp) obtained with static foams at high frequencies in a Bendix Ultraviscoson.

Observations of slip for foams in contact with a smooth surface were reported by Wenzel et al. in the case of a coaxial-cylinder viscometer [132]. The inner cylinder wall was roughened with vanes and the outer cylinder (rotor) wall was smooth. Besides, a modified cone-plate viscometer with both surfaces vaned was used to minimize slip effects. Extrapolating data of wall shear stress vs. rotational velocity, values of yield stress were reached below which, no flow occurred for a variety of dry foams of different texture and quality (>0.97).

2.3.1.2 Viscosity of Foams in Porous Media

Most of the researches on foams in porous media are concerned with the apparent viscosity of foams. To use apparent viscosity of foam to describe its behavior in porous media has been suggested by Heller et al. [119]. They suggested that the simple ratio between steady-state superficial velocity and pressure gradient (mobility), would better describe the nature of the flow.

As mentioned before, it is widely recognized that the apparent viscosity of foam is geometry dependent. It has been demonstrated both theoretically and experimentally [127], that apparent viscosity will decrease by a factor of 10^4 as the ratio of bubble diameter to tube diameter increases from 10^{-1} to 20 or so. It has been pointed out [134, 129] that foam with much smaller

cells than the pore throats of a porous media would have a very low mobility. Although such a situation would arise in certain cases, it is usually intended to reduce the mobility of a displacing fluid by a much smaller amount of gas.

2.3.1.3 Viscosity of Foams in Capillary Tubes

Several investigators have used continuous flow, tube viscometry [125, 128, 131, 132, 136] to evaluate the rheology of foam. In most studies, foam which is externally generated is passed through capillary tubes. The motivation to use small diameter tubes is to increase the applicability of such results to porous media.

In flow through long tubes or channels there would be a significant difference in foam density. This is due to different compressibilities of the component fluids in foam. Three consequences of this cell expansion in foams are as follows. (1) The bubble-size distribution of foam will change with the distance along the tube. This tends to make the geometry effect more pronounced (2) Foam quality will increase at the exit. This will affect foam flow substantially. (3) When the foam is assumed to be compressible, conventional viscometric analysis cannot be applied [129]. These phenomena will influence the flow mechanism of foam, which will make the reported viscosity values to be highly dependent upon the dimensions of the tubes used.

To explain the dependence of apparent foam viscosity to the tube radius, slippage at the tube wall has been used [136]. However, since the slip-corrected viscosity still retained a dependence on tube diameter the effort was not entirely successful. Dependence of apparent foam viscosity to the tube radius was also observed by others [125, 127, 128, 131]. Theoretical and experimental work of Hirasaki and Lawson indicate that in small-diameter capillary tubes foam texture is an important parameter [127]. Steady-state flow experiments for foams of different texture through two round tubes (one smooth and one rough) and three rectangular channels have

been performed by Wenzel et. al. [135]. The data were not used to calculate an apparent viscosity, but a finite slip velocity was reported for foam flow in smooth conduits.

2.3.2 Viscosity and Friction Factor of Bubbly Flow

It is known from theoretical analysis [137-139], laboratory experiments [140-146], and numerical simulations [147, 148] that rheological properties of suspensions are affected by bubbles. Despite this there has, until recently, been very little effort to include bubble suspension rheology in numerical flow models (but see Mastin [149]). This is historically due to two deficiencies in the literature: 1) researches on viscosity of bubble percentage, the other is that viscosity decreases with increasing bubble percentage; 2) each group has been unsuccessful in reaching an agreement on the relationship of viscosity and bubble percentage. There have been investigations conducted on the sign of the apparent viscosity: bubbles are capable of either increasing or decreasing the shear viscosity of a suspension depending on the dynamic regime [143, 145, 146, 150]. Each of these studies has also added new experimental data to understand the functional form of viscosity.

2.3.2.1 Viscosity of Bubble in Bulk

In order to understand the rheological behavior of bubbles, rotational viscometers can be used [15]. In these viscometers shear can be independently controlled and they can be used to study non-Newtonian behavior. One type of the rotational viscometers applied, measure the apparent shear viscosity with a wide-gap parallel plate rheometer by imposing a simple shear flow. Lim et. al. have used this type of viscometer to measure the viscosity of microbubbles [153151]. They have assumed that since their maximum bubble size has been 500 μm , bubble rising during the experiments can be ignored.

In some other cases the viscosity of bubble has been measured using the Stokes drag of a falling sphere in falling sphere viscometry [152]. Bubble's transient deformation can be evaluated in this method. In this method viscosity is directly obtained by the terminal velocities of the falling sphere relative to single-phase fluid.

In some of the previous literatures, researchers have tried to investigate bubble suspension's viscosity by theoretical analysis. Seo and Youn proposed a theoretical model to investigate rheological behavior of bubble suspension with large deformation [153]. In their theoretical model, constitutive equations for dilute bubble suspensions are derived by applying a deformation theory of ellipsoidal droplet. In another research Llewellin et. al. developed a semiempirical constitutive model for the visco-elastic rheology of bubble suspensions with gas volume fractions <0.5 and small deformations [146]. The constitutive equation takes into consideration, observable material parameters: the viscosity of the continuous phase, gas volume fraction, the relaxation time, bubble size distribution and an empirically determined dimensionless constant.

2.3.2.2 Viscosity of Bubbles in Porous Media

There have been limited researches done on bubble flow in porous media. To pass bubbles through pore throats the bubbles need to be considerably smaller than the pore throats of the porous media. Therefore, microbubble flow in porous media has gained a great interest due to the small size of microbubbles. Samuel et. al. have injected aqueous microbubbles, as a fluid with reduced mobility [14]. In their experiments, they have measured the oil recovery and the flow behavior of microbubbles.

Wan et.al. have conducted experiments to examine microbubble transport properties in porous media in order to evaluate their possible use in subsurface remediation [8]. After testing breakthrough of microbubble for different physical and chemical parameters of suspensions they

have developed a mathematical model. Their model is a special case of filtration theory to predict microbubble transport in porous media.

Choi et. al. conducted experiments of microbubble flow in porous media in a visual bed [154]. They observed that the microbubble suspensions were separated into a liquid and gas phase directly after injection. This showed much faster movement of liquid phase flow. They also observed that the gas front of the microbubble suspensions flowed in a plug-flow manner. This property of microbubbles in porous media can be effective in oil recovery and mobility control to eliminate instability of microbubble front.

2.3.2.3 Viscosity of Bubbles in Capillary Tubes

There have been numerous investigations on the effect of bubbles on wall friction and on bubble distribution in the cross section of the flow in tubes [156-162]. However, most of these studies focus on either large bubble sizes (0.5-2 *mm*) or flows in vertical tubes. It has been indicated by several investigators that the presence of bubbles results in a flattened liquid velocity at the center of the tube [156-159]. This behavior is caused by the bubble void fraction distribution along the cross section. It is shown that bubble distribution reaches its maximum close to tube wall. This maximum is more pronounced as the continuous phase Reynolds number

$$(\text{Re}_{c} = \frac{\rho_{c} vD}{\mu_{c}})$$
 reaches values under 5000 (Re_c<5000). In the case of higher Re_c values, the

suspensions behave as a homogenous flow [160]. These investigators have also demonstrated that, as the bubble size decreases, the bubbles tend to migrate closer to tube walls. Presence of bubbles close to tube walls result in an increased friction to the flow.

All of these studies have focused on bubble size in range of 0.5-2 *mm*. Therefore, investigations conducted in horizontal tubes have resulted in a partial separation of bubbles from the flow. The separation has caused a denser collection of bubbles on the top part of the tube [161]. This

behavior has been more pronounced in the case of low Re_c values. There have also been investigations done on microbubbles in horizontal tubes. Chernyshev investigated microbubble (mean diameter = 40 μ m) behaviors in tubes; the effect of microbubbles on the wall friction factor in tubes with diameters of 1.8 and 2.9 *cm* was considered [162]. It was found that the increase in wall friction factor was only a factor of the Re_c and the bubble void fraction. To our knowledge, investigations have not yet been conducted to relate the viscosity of microbubbles to the wall friction factor in very small Re_c (Re_c<50) in capillary tubes. Moreover, the viscosity of microbubble suspensions in capillary environments is one of the most important parameters involved in understanding their behavior.

In this work, the viscosity of microbubbles is measured in capillary tubes using a pressure drop along the tubes. Experimental wall friction factor has also been calculated by the measured pressure drops. The effect of tube diameter, tube length, microbubble void fraction, microbubble size distribution, and the Re_c on the rheological properties of microbubbles is taken into consideration. Subsequently, the experimental data have been used to develop a correlation to predict viscosity in capillary tubes. Besides, the relationship between wall friction factor and non-Newtonian Reynolds number (Re_n) has been investigated.

Chapter Three: Experimental

3.1 Microbubble Generator

Microbubble generators trap gas into a liquid using jet entrainment, cavitation or turbulence at the gas-liquid interface to entrain bubbles into the liquid. While some methods are capable of generating more uniform bubbles, others are easier to operate and are used for bulk production of microbubbles. In this study, the apparatus introduced by Sebba [88] was used with some modifications, as shown in Fig. 10.

The main part of the generator is the impeller, which is surrounded by a perforated casing that functions as the baffles. Air bubbles entrain into the water when agitations created by the impeller move through the perforations. The spinning impeller was mounted 5cm below the liquid surface. The speed of the impeller was found to have an impact on microbubble size distribution and stability. At low spinning speeds, the generated bubbles showed low stability. On the other hand, high spinning speeds created very small bubbles not suitable for micro studies. In this work, 13000 r.p.m. was the optimum spinning speed, generating the optimum bubble size with a good stability.

As mentioned by Save et al. spinning times below a certain value could affect the stability of the generated bubbles [6]. The effect of spinning time was investigated to find the threshold after which the time of spinning did not affect the stability of bubbles. It was found that 5 minutes of mixing would generate bubbles with good stability. Mixing times of more than 5 minutes have not shown an improvement on stability. The bubbles were found to remain stable for 12 hours, which was adequate to complete a series of experiments.



Fig. 10 Microbubble generator (modified high speed mixer based on the apparatus of Sebba [88])

3.2 Materials

In this study, air was used as the gas phase and a mixture of hydrophobic and a hydrophilic surfactant in deionized (DI) water was used as the liquid phase. The reason to select air as the gas phase is twofold: firstly, because of the ease of access to air and, secondly, to minimize gas phase solubility in the liquid phase and phase behavior.

A mixture of 1 g/L sorbitan monostearate (Span 60) and 1 g/L sodium dodecyl sulfate (SDS) was used to include the hydrophobic and hydrophilic surfactants, respectively. This concentration of both surfactants has been reported by Wan et al. to form the most stable microbubbles [8]. To create stable microbubbles, both of the surfactants need to be dissolved in water. Since Span 60 is a hydrophobic surfactant, it hardly dissolves in water. For its complete dissolution in water, it was necessary to leave the mixture for 24 hours to let the Span 60 adsorb water. The surface tension of the mixture was obtained by the pendant drop method and determined to be 44.3 *dyne/cm*. Moreover, the viscosity of the mixture was measured to be constant 0.99 *cp* at different shear rates. The density of the mixture was found to be equal to 1.03 g/cm^3 .

3.3 Microbubble Flow Test Setup

A schematic diagram of the setup employed in the experiments is shown in

Fig. 11. The main element of the setup is a circular glass tube (PYREX Brand Glass Tubing). Tube diameters of 0.47, 1.04 and 1.37 *mm* were used. As tube diameter has an extensive effect on calculations, the tube diameters were measured by taking pictures of a cross section of the tubes by a microscope (Scioptic LTM-402) with a maximum magnification power of 1000X. Tube lengths of 0.75, 1.00 and 1.22 *m* were employed to see the effect of tube length.

Both ends of the glass tube were connected to three way plexiglass chambers. The injection chamber was connected to the injection lining, transducer and the tube. The effluent chamber

was connected to the effluent lining, transducer and the tube. During pressure measurements, it was confirmed that there was no air trapped in the chambers in order to prevent errors. In the lining connecting the glass tube to the transducer several valves were employed. The purpose of the valves was twofold: first, to protect the transducer from over range pressures and, second, to calibrate the transducer (which will be explained more later).

The injection of the microbubbles into the capillary tube was carried out by a pulse-free constant flow syringe pump (Teledyne ISCO 500D). The microbubble suspension was first transferred to a transfer cylinder and then injected from the transfer cylinder at an accurate constant flow rate using the syringe pump. Before the injection of the microbubble suspension, de-ionised water was circulated through the setup to fill the teflon tubes with water, in order to eliminate the presence of microbubbles in the connection lines, the presence of which may affect the pressure measurement.



Fig. 11 Schematic diagram of capillary tube setup

3.4 Procedure

To measure the viscosity of the microbubble suspension through the capillary tube, the differential pressure was measured using a high-sensitive transducer (DP103, Validyne Engineering). Electric signals transmitted by the transducer were switched to digital signals by a demodulator (Validyne CD15) so as to be readable by the computer software (Instacal Version 5.52). For the comprehension of the signals, the transducer should first be calibrated using water column. Calibration was performed by reading the signals at three different water column heights. The line connecting these three points provided the equation to convert signals to differential pressures.

Pressure drops of microbubble suspensions were measured for different tube diameters, tube lengths, flow rates, and gas void fractions. All of the pressure drop measurements were conducted at a constant temperature (21°C). The effect of each parameter was investigated by keeping the other variables constant. Beside using the aforementioned tube diameters and tube lengths, four gas void fractions of 3.4, 4.9, 6.4 and 7.8% were used to study the effect of gas percentage on the viscosity of microbubble suspensions. Furthermore, to examine the dependence of viscosity on flow rate, the pressure drop was measured in different flow rates while keeping the other variables constant. Since the effect of flow rate was investigated through Re_c , flow rates examined were carefully chosen to yield equal Re_c numbers in different tube diameters.

To have a better understanding of microbubble deformation through capillary tube differential pressure measurements, the capillary number was measured ($Ca = \frac{\mu_c \gamma a}{\sigma}$). In this study, Ca was varied from 4.6×10^{-6} to 3.8×10^{-4} depending on the bubble size distribution. As mentioned by Pal for small values of Ca, close to zero (Ca < 0.01), microbubbles can be assumed to be

undeformable [163]. Therefore, in this study, due to Ca numbers close to zero, microbubbles are assumed undeformable.

Chapter Four: Results and Discussion

4.1 Analysis of Experimental Data

4.1.1 Determination of Viscosity of Microbubbles

To calculate the experimental viscosity of microbubble suspensions, measured pressure drops were used. In this method, the theoretical expression given by Rabinowitsch was used to calculate the rate of the shear to microbubbles [164]. This theoretical analysis was also presented by Mooney [165] and Metzner and Reed [166]. It has also been applied to measure viscosities of liquid-liquid emulsions by Alvarado and Marsden Jr. [167]. The expressions are as follows:

$$\tau = \frac{r\Delta P}{2L} \tag{1}$$

$$\tau = K' \left(\frac{8\nu}{D}\right)^{n'} \tag{2}$$

$$\gamma = \frac{3n'+1}{4n'} \left(\frac{8v}{D}\right) \tag{3}$$

where τ and γ are shear stress and shear rate at the capillary wall, respectively. Capillary radius, diameter and length are denoted by *r*, D and *L*. ΔP is the pressure drop along the capillary tube and *v* is the average flow velocity. Eq. 1 is used to calculate shear stress. Since Eqs. 1 and 2 are both expressing shear stress, one can write:

$$\frac{r\Delta P}{2L} = K' \left(\frac{8v}{D}\right)^{n'} \tag{4}$$

According to Eq. 4, n' is the slope of the line drawn to represent $r\Delta P/2L$ versus 8v/D on a logarithmic plot. Thus, n' can be calculated as follows:

$$n' = \frac{d \ln\left(\frac{r\Delta P}{2L}\right)}{d \ln\left(\frac{8v}{D}\right)}$$
(5)

After obtaining *n'*, *K*['] can be found as the y-intercept of the line drawn representing $r\Delta P/2L$ versus 8v/D on a logarithmic plot. It has been shown that *n*['] is constant over a wide range of shear stress for non-Newtonian fluids [166].

By definition viscosity is known as:

$$\mu_a = \frac{\tau}{\gamma} \tag{6}$$

Substituting Eqs. 2 and 3 into equation 6 results in the following equation, which is used to calculate the apparent viscosity of microbubble suspensions.

$$\mu_a = \frac{4n'}{3n'+1} K' \left(\frac{8\nu}{D}\right)^{n'-1}$$
(7)

The apparent viscosity calculated from Eq. 7 is the experimental viscosity of microbubbles obtained from the pressure drop in capillary tubes.

4.1.2 Determination of wall friction factor of Microbubbles

One of the main parameters affecting fluid flow in tubes is friction factor. In capillary tubes the boundary layer close to wall is comparable to the cross section of the tube in capillary tubes. Therefore, effect of wall friction on the flow gets more pronounced in capillary tubes.

In order to calculate wall friction factor, pressure drop has been utilized in this study. The method used to calculate friction factor has been adopted from the studies of Metzner and Reed [166]. In this method pressure drop has been applied to the following equation:

$$F = \frac{r\Delta P}{2L} \left/ \frac{\rho v^2}{2} \right.$$
(8)

where F is the friction factor. Equation 8 can be simplified as follows:

$$F = \frac{r\Delta P}{L\rho v^2} \tag{9}$$

4.2 Rheological Behavior of Microbubbles

The calculated n' from Eq. 5 represents the flow behavior index, which demonstrates the rheological behavior of microbubbles. In Fig. 12 to 14, $r\Delta P/2L$ versus 8v/D are shown in *Ln-Ln* plots [176]. Fig. 12 and 13 show the plots of different tube diameters and lengths, respectively. Fig. 14 presents the lines of different bubble void fractions. According to Eq. 5 the slopes of the lines show n' for each series of data. Therefore, lines with the same slopes are representing suspensions with similar rheological behavior. The lower the value of the slopes of the lines is from one, the more the suspensions deviate from Newtonian behavior.



Fig. 12 $r\Delta P/2L$ versus 8v/D for different tube diameters, constant tube length and constant bubble void fraction ($\phi = 4.9\%$)



Fig. 13 $r \Delta P/2L$ versus 8v/D for different tube length, constant tube diameter and constant bubble void fraction ($\phi = 4.9\%$)



Fig. 14 *r*Δ*P*/2*L* versus 8*v*/*D* for different bubble void fractions, constant tube diameter and length (*D*=1.04 *mm L*=1.22 *m*)

In Fig. 12 and 13, although the value of the viscosity changes in different tube diameters and lengths, the slopes of the lines are constant (0.69) (Table 1). This is due to a constant bubble void fraction in suspensions. However, in Fig. 14, the slope of the lines changes in different bubble void fractions (0.62 to 0.72) (Table 1). This suggests that tube conditions may affect viscosity, but the rheological behavior is determined by the percentage of bubbles in the suspension.

4.3 Correlation of Microbubble Viscosity

In this study, the model proposed to predict the viscosity of microbubble suspensions in capillary tubes is started from Poiseuille's formula [168]:

$$\frac{\Delta P}{\rho_c v^2} = \frac{16}{\text{Re}_c} \left(\frac{L}{r}\right)$$
(10)

where ρ_c is the continuous phase density. Poiseuille's formula has been defined for fully developed laminar flow with a parabolic velocity profile. It has been suggested that the pressure drop in capillary tubes for single phase liquids is higher than the value predicted by Poiseuille's formula [168]. There are two main reasons for this difference in the pressure drop of a single phase flow in capillary tubes and the predicted value of Poiseuille's formula. First, in the initial part of the flow, the profile is changing from a near-uniform distribution to a parabolic velocity distribution. This will cause an additional pressure drop compared to Poiseuille's formula. Second, the acceleration at the centre of the tube in the initial region of the flow causes a radial velocity which leads to a radial pressure drop. This additional pressure drop will result in an underestimation of Poiseuille's formula, analytically, by simplified mathematics [169-170]. Langhaar proposed a modified equation for Re_c $\rightarrow \infty$ [169]. This equation was achieved by linearizing Navier-Stokes equations. The proposed equation is as follows:

$$\frac{\Delta P}{\rho_c v^2} = \frac{16}{\text{Re}_c} \left(\frac{L}{r}\right) + 2m \tag{11}$$

where *m* is experimentally determined. Other investigators have applied numerical solutions to solve the problem [171-175]. All of the solutions present *m* in Eq. 11 as a function of Re_c . Therefore, Eq. 11 can be written as:

$$\frac{\Delta P}{\rho_c v^2} = \frac{16}{\text{Re}_c} \left(\frac{L}{r}\right) + m(\text{Re}_c)$$
(12)

Kestin et al provided a correlation for *m* [168]. Their study was done for $0.5 < \text{Re}_c < 100$. They concluded that their correlation could also be applied to $\text{Re}_c > 100$, since the values of their correlation in Re_c exceeding 100 were close to values presented by Langhaar [169]. They correlated *m* as:

$$m = m_0 + \frac{8n}{\text{Re}_c} \tag{13}$$

 m_0 and *n* are constants, which need to be found experimentally. To reach the viscosity, they introduced the proposed equation (Eq. 13) into Eq. 12, which resulted in:

$$\mu_a = \frac{\pi r^4 \rho_c}{8(L+nr)} \left(\frac{\Delta P}{\dot{m}} - \frac{m_0 \dot{m}}{\rho_c \pi^2 r^4} \right)$$
(14)

Eq. 14 uses a virtual length instead of the actual tube length. The virtual length (L' = L + nr) corrects Poiseuille's formula, creating a better prediction of capillary tube pressure drop and viscosity. In their study m_0 and n were found, theoretically, to be 1.17 and 0.69 respectively.

It has been found by Kestin et al that the behavior of fluids in capillary tubes is sensitive to the inlet conditions [168]. The inlet conditions include both the shape of the tube at the entrance and the size of the injection reservoir, compared to the capillary diameter. In this study, in order to eliminate the effect of the injection reservoir size, the plexiglass chambers used to inject microbubbles into the tube were chosen to be considerably larger than the tube diameter. Furthermore, capillary tubes were square-cut at the entrance, so as to have a sharp inlet for the flow.

In this study Eq. 14 was tried to predict the viscosity of microbubble suspensions. It was found that Eq. 14 was capable of predicting microbubble viscosities in a constant bubble void fraction

by changing the coefficients (m_0 and n). However, when the bubble fraction was changed, Eq. 14 was unable to predict microbubble viscosities. Therefore, L' was modified in order to obtain a better prediction [177]. In this study L', which was defined as L+nr in Eq. 14, was modified for different bubble void fractions as follows:

$$L' = \left(L + nr\right) \left(\frac{\phi}{\phi_r}\right)^x \tag{15}$$

where x is a constant that is found experimentally and ϕ_r is the reference gas void fraction, which is the bubble percentage in which m_0 and n are determined. Thus, Eq. 14 was modified to:

$$\mu_{a} = \frac{\pi r^{4} \rho_{c}}{8(L+nr)\left(\frac{\phi}{\phi_{r}}\right)^{x}} \left(\frac{\Delta P}{\dot{m}} - \frac{m_{0}\dot{m}}{\rho_{c}\pi^{2}r^{4}}\right)$$
(16)

Parameters m_0 , n and x need to be found through an optimization process. In order to find these parameters, the data from a constant bubble fraction (ϕ_r) were used to reach the parameters in Eq. 14 (m_0 and n). After obtaining m_0 and n, the data from different gas void fractions were applied to Eq. 16 in order to reach x. To perform optimization, half of the data was selected randomly. After finding the parameters, the other half of the data was used to verify the model with the fitting parameters.

In the introduced correlation (Eq. 16) viscosities of microbubbles can be predicted using their bubble void fraction. As mentioned before, although the value of viscosity changes with tube conditions, the rheological behavior of microbubbles is dependent on the bubble void fraction. In other words, n' only depends on bubble void fraction. Therefore, in Eq. 7, which is used to calculate experimental viscosity, the effect of bubble void fraction is presented by n'. To

accommodate the effect of the bubble void fraction on apparent viscosity in the proposed equation (Eq. 16), the modification introduced in Eq. 15 ($\left(\frac{\phi}{\phi_r}\right)^x$) was implemented.

4.4 The Viscosity of Microbubbles in Capillary Tubes

The results for the viscosities of microbubbles in different tube diameters and lengths are shown in Fig. 15 and 16. Moreover, the results for different bubble void fractions can be found in Fig. 17 [177].

Fig. 15 to 17 illustrate the relationship of viscosity and Re_c . When Re_c increases, the viscosity of microbubbles decreases. The viscosity decreases until it reaches a constant value. Increasing Re_c after this point results in a constant viscosity of microbubbles. This represents the non-Newtonian behavior of microbubbles in the capillary tubes. It is known that small bubbles (<0.6mm) start to concentrate close to the tube walls upon their entrance into a tube [158, 159]. This causes the void profile of the gas to demonstrate two maximus close to the tube walls and a constant value at the centre of the tube. By increasing Re_c , the bubble distribution becomes more uniform in the centre of the tube and the two maximums close to walls start to diminish. As mentioned by Valukina et al for increased values of Re_c , the friction factor in tubes is decreased [158]. A decrease in the friction factor leads to a lower viscosity of the suspension and a lower pressure drop across the tube.



Fig. 15 Experimental results for microbubble viscosity in different tube diameters and constant tube lengths and void Fraction $(\phi = 4.9\%)$



Fig. 16 Experimental results for microbubble viscosity in different tube lengths and constant tube diameters and void Fraction $(\phi = 4.9\%)$


Fig. 17 Experimental results for microbubble viscosity in different bubble void fractions and constant tube diameters and lengths (*D*=1.04 *mm L*=1.22 *m*)

In Fig. 15, a decrease in viscosity with the decrease in tube diameters is observed. This was also caused by a different bubble orientation on the cross section of the tube. Smaller tube diameters lead to more uniform void distributions in a constant bubble concentration, which causes less pressure drop across the tube and smaller viscosity values.

Fig. 16 represents the effect of tube length on the viscosity of microbubbles. Smaller viscosities are observed in longer tubes. According to Kestin et. al. in capillary tube viscometry there is an additional pressure drop caused by the developing region at the beginning of the flow [168]. In this work, the effect on the viscosity of microbubbles was more pronounced than in single phase flow. The pronounced effect was caused by the disturbance of the developing region of bubble distribution, which was present in addition to the velocity profile developing region of the continuous phase. As the length of the tube increased, the additional pressure drop caused by the developing regions got smaller, compared to the overall pressure drop. Thus, the calculated viscosity decreased as the tube length increased.

Fig. 17 illustrates the effect of bubble void fraction on the viscosity of suspensions. As the fraction of the bubbles increased the viscosity increased. It has been reported that an increase in bubble void fraction increases the wall friction factor [162]. An increase in friction factor results in the increase of viscosity, which eventually leads to the increase in pressure drop of suspensions in capillary tubes.

Beside the bubble void fraction, the effect of bubble size distribution on the viscosity of the suspension was taken into consideration. To study the effect of size distribution, the viscosities of suspensions were measured in different bubble size distributions while keeping the quality of the suspensions constant. The pictures of suspensions and bubble size distributions are shown in Fig. 18 to 20 [177].

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Fig. 18 Microbubble photo and size distribution (Distribution 1)



Fig. 19 Microbubble photo and size distribution (Distribution 2)



Fig. 20 Microbubble photo and size distribution (Distribution 3)

The viscosities of different bubble size distributions have been measured in identical tubes to eliminate all of the affecting parameters except bubble size distribution. The viscosity results are shown in Fig. 21 [177]. The results illustrate similar viscosity values for different bubble size distributions in constant Re_c numbers. This represents the independency of suspensions' viscosities from bubble size distribution. As previously reported by Chernyshev [162], it is expected to have independency of suspensions' flow behavior from bubble size distribution in small bubble size ranges (<0.5 *mm*) [162]. The results in this study confirm the finding previously reported by Chernyshev [162].



Fig. 21 Experimental results for microbubble viscosity in different bubble size distributions with a constant bubble void

fraction (Tube: D=1.04 *mm* L=1.22 *m* ϕ =6.4%)

Equation 16 was used to obtain the experimental viscosity data. In this work, m_0 , n and x were found to be equal to 1.25, 207.1 and 0.024, respectively [177]. Compared to single phase studies performed by previous investigators, m_0 is in the same order. However, n, which defines the virtual length, has increased considerably. This is due to the presence of microbubbles, which causes a higher pressure drop as compared to a single phase fluid. This has caused the virtual length to be higher than in single phase studies in order to represent the extra pressure drop.

Fig. 22 to 24 show the comparison between the experimental relative viscosity and the relative viscosity results from the proposed correlation [177]. The reason for using relative viscosity in comparing the experimental and correlated results is to see the effect of bubbles on the viscosity of the suspension. In other words, the ability of the correlation in predicting the bubble effects on suspension's viscosity is shown in Fig. 22 to 24; this illustrates that the proposed correlation is capable of predicting the relative viscosity of microbubbles in tubes of different diameters (Fig. 22: D = 0.47, 1.04 and 1.37mm, L = 1.22 m, $\phi = 4.9\%$) and different lengths (Fig. 23: L=0.75, 1.00 and 1.22 m, D = 1.04 mm, $\phi = 4.9\%$) and different bubble void fractions (Fig. 24: $\phi = 3.4$, 4.9, 6.4 and 7.8%) in the range of Re_c<50, with AARD under 1.3%. The maximum deviation between the experimental data and the proposed correlation results was equal to 2.6%.



Fig. 22 Experimental data versus proposed correlation results for different tube diameters at a constant tube length and bubble void fraction (ϕ =4.9%)



Fig. 23 Experimental data versus proposed correlation results for different tube lengths at a constant tube diameter and bubble void fraction (ϕ =4.9%)



Fig. 24 Experimental data versus proposed correlation results for different bubble void fractions at a constant tube diameter and length (*D*=1.03 *mm L*=1.22 *m*)

4.5 The Wall Friction Factor of Microbubbles in Capillary Tubes

It is known form Poiseuille's formula that for single phase flow in laminar condition, friction factor exhibits a linear relationship with Reynolds number as follows:

$$F = \frac{16}{\text{Re}} \tag{17}$$

Metzner and Reed [166] proposed that replacing conventional Re with non-Newtonian Reynolds number will make Eq. 17 capable of describing friction factor for non-Newtonian fluids. It has been indicated that, non-linear relation of friction factor and non-Newtonian Re in non-Newtonian fluids demonstrates either errors in the experiments or thixotropic or rheopectic behavior of the fluid. They defined non-Newtonian Re by replacing viscosity in Reynolds number with the apparent viscosity of non-Newtonian fluids. Therefore applying apparent viscosity (Eq. 7) to Re will result in:

$$\operatorname{Re}_{n} = \frac{D^{n'} v^{2-n'} \rho}{K' 8^{n'-1}}$$
(18)

Metzner and Reed [166] described the relation between Re_n and F as follows:

$$F = \frac{16}{\text{Re}_n} \tag{19}$$

Previous studies on friction factor of bubbles in tubes have been conducted on large sizes of bubbles or tubes [158, 160]. It has also been done on friction factor of microbubbles. Serizawa conducted experiments on microbubbles with bubbles diameter of 40μ m in vertical acrylic tube of 20mm inner diameter [178]. He explained friction factor with continuous phase Re. The same experiments have been conducted by Chernyshev with same bubble diameters [162]. However, his experiments were conducted in horizontal tubes. He also described friction factor of microbubbles with continuous phase Re_c.

In this study the relation between non-Newtonian Re_n and friction factor for microbubbles in capillary tubes in laminar conditions have been investigated. Since it has been explained in the rheological behavior section that microbubbles present non-Newtonian behavior, friction factor is explained by non-Newtonian Re_n .

First friction factor of microbubbles was calculated through Eq. 9. Then they have been illustrated in Fig. 25 [177].

Fig. 25 demonstrates calculated friction factors from experimental data versus non-Newtonian Re_n. Non-Newtonian Re_n has been calculated using the flow indexes achieved from Fig. 12 to 14. Fig. 25 shows linear relation between experimental friction factor of microbubbles and non-Newtonian Reynolds number [177]. A very good agreement between the predicted results from the proposed formula for friction factor of non-Newtonian fluids (Eq. 19) and the experimental data was achieved. Therefore it is concluded that the formula proposed by Metzner and Reed for friction factor in non-newtonian fluids (Eq. 19) can be applied to friction factor of microbubbles in capillary tubes.



Fig. 25 Friction factor versus non-Newtonian Reynolds (Re_n) number for laminar microbubble flow

Symbol	Tube Diameter,	Tube Length, m	Bubble Void	Flow Index, <i>n'</i>
	mm		Fraction, %	
0	0.47	1.22	4.9	0.693
$\square \square \square$	1.03	1.22	4.9	0.693
	1.36	1.22	4.9	0.690
\bigcirc	1.03	1.00	4.9	0.699
\bigcirc	1.03	0.75	4.9	0.698
*	1.03	1.22	3.4	0.721
+	1.03	1.22	6.4	0.655
	1.03	1.22	7.8	0.619

Table 1 Flow index of microbubbles in different tube condition and bubble void fraction

Chapter Five: Conclusions and Recommendations

In this study, experiments were conducted to determine microbubble viscosity and wall friction factor in capillary tubes. The pressure drop of the microbubbles was measured in three different tube sizes and lengths. Moreover, pressure drops of microbubbles in different bubble void fractions and with different bubble size distributions were measured. Pressure drops were utilized to calculate experimental viscosity and wall friction factor.

5.1 Conclusions

The microbubbles presented non-Newtonian behavior. When Re_c was increased, the viscosity of microbubbles decreased. The viscosity decreased until it reached a constant value. Increasing Re_c after this point resulted in a constant viscosity of microbubbles.

The viscosity of microbubbles decreased as the tube diameter decreased and the tube length increased. This reduction in viscosity was due to different bubble orientations in the cross section of the tube. Furthermore, the viscosities of the suspensions increased with an increase in the bubble void fraction. The increase in viscosity because of an increase in the void fraction of bubbles coincides with a change in the flow behavior index (n). However, the change in the viscosities of microbubbles due to a change in capillary tu be length and diameter did not affect the flow behavior index. These results showed that although tube conditions change viscosity values, they do not affect the rheological behavior of suspensions. The only parameter that affects the flow behavior index is the void fraction of bubbles. The effect of bubble size distribution was also investigated. It was found that, in the bubble size range of 1-12 μm , the size distribution of bubbles did not influence the viscosity of suspensions.

Finally, data obtained from experiments were used to develop a correlation to predict the viscosity of microbubbles flowing through capillary tubes. A good agreement between experimental viscosity data and correlated results was obtained. The developed correlation is capable of predicting microbubble viscosity in the range of $Re_c < 50$.

Experimental wall friction factor was calculated from obtained pressure drops. Re_n has also been calculated using the flow index of microbubbles. It has been observed that the relationship between wall friction factor of microbubbles and Re_n follow the same formula for non-Newtonian fluids presented by previous researchers.

5.2 Recommendations

Based on the results obtained in this research, there are several future paths that seem promising and can be investigated:

5.2.1 Microbubbles in Porous Media

This research has been conducted with the interest to simulate microbubble flow in porous media with the results obtained from capillary tubes. Since the results in this research show an increase in viscosity of microbubbles in capillary tubes, it is relevant to recommend microbubbles for mobility control problems in enhanced oil recovery. However, for microbubbles to be successfully applied to porous media there are several issues that need to be addressed:

1) To make sure the size of bubbles are suitable for the flow of bubbles in the pores of porous media. Otherwise, the porous media will act as a filter to bubbles. Continuous gas phase is formed in this case and bubbles will not be effective in mobility control.

2) Microbubbles need to be stable enough to prevent them from being destroyed. Since microbubbles require higher pressure than single phase fluids to flow through porous media, they must be able to tolerate the pressure applied to them.

5.2.2 Microbubble Flow Involving Phase Behavior

Most of the applications of microbubbles involve phase behavior during their flow through porous media. Therefore, it is recommended to expand the result from this research to flows with phase behavior involved in them. There are two preferred systems that can be recommended for future experiments due to their high application:

1) Microbubbles of CO_2 in water can be examined in porous media. Due to the solubility of CO_2 , these microbubbles will change size during their flow through porous media. Environmental motivations have made CO_2 microbubbles a good candidate for carbon sequestration. Evaluation of CO_2 microbubbles' behavior in porous media will be of great assistance for carbon sequestration.

2) Hydrocarbon microbubbles produced in result of pressure reduction in heavy oils are another good candidate for future researches. These microbubbles have been reported to have reduced viscosity compared to single phase heavy oil. However, researches done on this matter have not been in agreement with each other. Thus, they have been inconclusive. It is recommended to primarily conduct hydrocarbon microbubble flows in capillary tubes to have solid evidence of the behavior of this type of microbubbles in capillary environments and porous media.

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