Dynamics and Rheology of Sheared Two-dimensional Foam

by

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A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF

DOCTOR OF PHILOSOPHY

 in

The Faculty of Graduate and Postdoctoral Studies

(Chemical and Biological Engineering)

THE UNIVERSITY OF BRITISH COLUMBIA

(Vancouver)

February 2014

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Abstract

Using a shear cell device, we have studied four associated problems in foam by experiments: Bubble-bubble coalescence in sheared two-dimensional foam; lateral migration of a single large bubble in an otherwise monodisperse foam; size segregation of bubbles in sheared bidisperse foam; and the effect of non-Newtonian rheology of foam on lateral migration of bubble. For bubblebubble coalescence in sheared two-dimensional foam, we observed a threshold of shear rate beyond which coalescence of bubbles happens. The most promising explanation was the model based on the centripetal force with qualitative agreement with experimental results.

Next we studied the dynamics of monodisperse foam in the presence of a single bubble whose size is different from the neighboring bubbles. We reported the lateral migration of a larger single bubble away from the wall. We also reported thresholds of shear rate and bubble size ratio beyond which migration occurs. In this study we modified the Chan-Leal model and predicted the experimental trajectories of migrating bubbles.

For bidisperse foams, we reported evolution in foam structure to a size segregated structure, in which large bubbles accumulate at the middle of the gap whereas smaller ones close to walls. Then, we adopted a model based on convection-diffusion equation to account for both lateral migration and shear induced diffusion.

Finally, we extended the second work by widening the gap of Couette coaxial cylinder geometry. Similar to the second work, we found that large bubble migrates laterally to an equilibrium position close to the inner wall. We believe this new mechanism is the non-Newtonian feature of foam. We characterized our foam by measuring its degree of shear thinning and also estimated its elasticity based on the literature data on foam. Then, we found out for a shear thinning fluid bubble migrated to position even closer to the inner wall than in the foam while a bubble in Boger fluid migrated to a position closer to the outer cylinder. Therefore, for a viscoselastic fluid which has the same feature one would expect to see bubble migration to a position between these two for two fluids.

Preface

This PhD thesis entitled "Dynamics and Rheology of Sheared Two-dimensional Foam" presents the main features of the research that I carried out during my PhD study under supervision of Professor James. J. Feng. In this preface, the contributions and collaborations to the papers published or submitted for publication from current thesis are briefly explained.

- A version of chapter 3 has been published. H. Mohammadigoushki, G. Ghigliotti, and J. J. Feng (2012), Anomalous coalescence in sheared two-dimensional foam. Physical Review E 85, 066301 (2012). Under supervision of J. J. Feng, and collaboration with Giovanni Ghigliotti, I did a comprehensive experimental work with theoretical study of coalescence in sheared two-dimensional foam and drafted the paper. J. J. Feng put his ideas and helped me to prepare the final version of paper.
- A version of chapter 4 has been published. M. H. Mohammadigoushki, and J. J. Feng (2012), Size-differentiated lateral migration of bubbles in Couette flow of two-dimensional foam. Phys. Rev. Lett. 109, 084502. Under supervision of J. J. Feng, following first study I performed experiments on cross stream-line migration of a single large bubble inside a monodisperse foam in a Narrow gap Couette device and drafted the paper. J. J. Feng helped me to explore the effect of different parameters in experiments as well as digging more into physics of the problem and he also helped me to prepare the final version of paper.
- A version of chapter 5 has been published. H. Mohammadigoushki, and J. J. Feng (2013), Size segregation in sheared two-dimensional polydisperse foam. Langmuir 29, 1370-1378. Through a systematic research, I studied the size-based segregation of bubbles in bidisperse and polydisperse two-dimensional foam and developed a model to explain the experimental observation. I conducted this study under supervision of J.J. Feng. I prepared the draft of the paper with help of J. J. Feng.
- A version of chapter 6 has been submitted for publication. H. Mohammadigoushki, P. Yue and J. J. Feng (2013), Bubble migration in

two-dimensional foam sheared in a wide-gap Couette device: effects of non-Newtonian rheology. Through a systematic research, I studied the lateral migration of single large bubble in monodisperse twodimensional foam in wide-gap Couette co-axial cylinder device and explained the experimental observations both by theoretical models and simulation. Professor Pengtao Yue at Virgina Tech helped us to get the simulation results for migration of a bubble in shear thinning fluid. I conducted this study under supervision of J.J. Feng. I prepared the draft of the paper with help of J. J. Feng.

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Nomenclature

\mathbf{Symbol}	Units (SI)	Description
A	J	Hamaker constant
a	m	Film radius
a		Short axes of deformed bubble
a_1, a_2, a_3	m	Bubble size
Ca		Capillary number
Ca_e		Effective capillary number
С	mol/m^3	Bulk concentration of surfactant
D	m/sec^{-2}	Surfactant diffusivity
D		Deformation parameter
D_e		Dimensionless Deborah number
d	m	Gap width
F	N	Force
F_b	N	Bumping force
F_c	N	Capillary force
F_w	N	Stokes wall repulsion force
h_0	m	Initial thickness of film
h_c	m	Critical film thickness for rupture
K	$Pa.sec^n$	Consistency factor
k_B	$m^2 kg s^{-2} K^{-1}$	Boltzman constant
l	m	Long axes of deformed bubble
Ma		Dimensionless Marangoni number
M	N.m	Torque at the inner cylinder
n		Power law index
Pe		Dimensionless Peclet number
R	m	Bubble size
R_i	m	Radius of inner cylinder
R_o	m	Radius of outer cylinder
R_e		Reynolds number
R_g	J/molK	Universal gas constant
r	m	Bubble size
r_s	m	Characteristic size of surfactants
S		Dimensionless position
T	K	Absolute temprature
t_i	sec	Interaction time between two bubbles
t_d	sec	Drainage time of the liquid film between two bubbles

u	m/sec	Tangential velocity
v_i	m/sec	Radial velocity at the inner cylinder
v_m	m/sec	Migration velocity
y		Dimensionless coordinate across the gap
y'	m	Dimensional coordinate across the gap
N_1	Pa	First normal stress differences

$Greek \ symbols$

\mathbf{Symbol}	Units (SI)	Description
μ	Pa.s	Shear viscosity of the fluid
η	Pa.s	Shear viscosity of the fluid
ρ	$kg.m^{-3}$	Density of the fluid
σ	$N.m^{-1}$	Surface tension of the fluid
σ	$N.m^{-1}$	Shear stress of the fluid
σ_i	$N.m^{-2}$	Shear stress at the inner cylinder
σ_o	$N.m^{-2}$	Shear stress at the outer cylinder
σ_y	$N.m^{-2}$	Yield stress of the fluid
Ω	rpm	Rotational velocity
Ω_c	rpm	Critical rotational velocity for coalescence
$\dot{\gamma}$	sec^{-1}	Shear rate
au		Dimensionless time
Γ	$mole/m^2$	Surfactant concentration at the interface of bubble
Γ_0	$mole/m^2$	Equilibrium surfactant concentration at the interface
		of bubble
α		Coefficient of adsorption
$ au_v$	Pa	Shear stress
$ au_M$	Pa	Marangoni stress
δ	m	Thickness of boundary layer
κ		Bubble size ratio
κ_0		Threshold for bubble size ratio
Ω_0	rpm	Threshold for rotational velocity
Φ		Dimensionless force
Φ_1		Dimensionless force for the first innermost row of
		bubbles in foam
Φ_1, Φ_2, Φ_3		Average area fractions of the bubble species
ϕ_1,ϕ_1,ϕ_1		Local area fractions of the bubble species
λ		Dimensionless diffusion coefficient

Acknowledgements

I completed this PhD thesis with the help and support of my supervisor, Professor James J. Feng, during which he provided me with his excellent and thoughtful supports and encouragements. He showed me how to be more critic and to dig the problems thoroughly, and not to be disappointed when the hard times of research come. I also would like to thank Professor G. M. Homsy for giving me many valuable and insightful advices during my PhD studies.

Chapter 1

Introduction

1.1 Background and application

Aqueous foams are highly concentrated dispersion of bubbles inside a surfactant solution. Despite the fact that foams contain gas and liquid that are simple fluids, their dynamics can be quite complex [13]. This complex behavior finds unique applications in several industrial processes. For instance, their low density and high surface area make them good materials for flotation in which liquid foams are used to extract minerals from ore [40]. Gases such as steam, carbon dioxide and hydrocarbon gases are injected into oil reservoirs to increase the recovery of oil. These gases are much less dense and less viscous than the oil they attempt to displace, so they tend to migrate to the top of the reservoir, leaving most of the oil behind. Foams can help these gases to sweep oil reservoirs more efficiently [40]. Liquid foams are being used in daily life as well, in cosmetics and foods.

1.2 Basic elements of a liquid foam



Figure 1.1: Typical Structure of a liquid foam.

A liquid foam is made up of some distinct structural elements (bubbles, films, and Plateau borders). This elegant structure is illustrated beautifully in the images shown in figure 1.1.

Films: In liquid foam bubbles are pressed together but are separated by thin films. Although these are the most evident feature of the foam structure, they become significant only in stability of foam, since foams break because of film rupture.

Plateau borders: These are where films meet in threes along an edge. This region is a liquid-filled channel.



Figure 1.2: Elements of a liquid foam in a magnified structure.

1.3 Liquid foams, wet and dry

Foam structure can be characterized by different parameters including the foam quality and distribution of bubble size. Foam quality refers to the fraction of gas inside the sample of foam. With increasing of the foam quality, the shape of bubbles may change from spherical to polyhedral and foam transforms from wet to dry.

Fig.1.3 shows samples of a wet foam and a dry foam. Based on the size distribution of bubbles, foams can be monodisperse, bidisperse or polydisperse. Liquid foams can also be categorized in terms of dimensionality to 2D foam and 3D foam.

Two-dimensional liquid foams: A so-called 2D foam is a monolayer of bubbles. Depending on how the monolayer is confined on the top and the bottom, there are three common configurations, as illustrated in Figure 1.4.

Three-dimensional liquid foams: If the bubbles in foam are in contact with each other in three dimensions they form a three-dimensional foam.

1.4 Stability of liquid foams

1.4.1 Quasi-static porcesses

A quasi-static process in a foam is one in which the relaxation of the structure back to equilibrium is much faster than the time-scale at which the foam



Figure 1.3: Typical Structure of a dry foam (A) and a wet foam(B).

is perturbed. Foams are unstable materials and therefore, surface active agents or surfactants are added to the solution to stabilize them. Surfactant molecules have a hydrophobic and a hydrophilic part. When adsorbed to the gas-liquid interfaces, they reduce the surface tension and generate Marangoni stresses that would inhibit the tangential flow along the interface of bubbles and therefore, foam becomes more stable. It has been shown that the foam is constantly evolving as soon as it is created. This complicates the measurements and therefore, alters experimental trends. Several mechanisms result in foam structural evolution. The structural evolution may occur in mechanical equilibrium or under dynamic fields. These processes may act at the same time. At mechanical equilibrium, three processes may lead to structural evolution , which are:

i) Gravitational drainage: Due to the effect of gravity liquid flows vertically and accumulates at the bottom of the column; this is gravitational drainage and leads to thinning of the film between bubbles and eventually rupture of the bubble.

ii) Bursting of bubbles: Bubbles in foam may burst at the interface with free air. It has been shown that the burst of one bubble in a sample of static foam might trigger some avalanches of bursting of neighboring bubbles as well (Vandewalle et al. 2002).

iii) Coarsening or Ostwald Ripening. For polydisperse foams, the Laplace pressure in neighboring bubbles is different. In smaller bubbles



Figure 1.4: Typical Structure of a two-dimensional foams. (b) Liquid foam with gas-liquid boundary condition (bubble raft), (b) Two-dimensional foam with glass-liquid boundary condition and (c) two-dimensional foam with glass-glass boundary condition (Hele-Shaw cell).

this pressure is higher than that in larger ones. This causes the gas to diffuse from small to large bubbles through liquid films; this process is called coarsening or Ostwald ripening. Consequently, strong coarsening may lead to topological changes called T1 and T2 events. T1 is a neighbor switching event in which bubbles switch their neighbors to gain a lower energy level (Weaire & Hutzler 1999). A typical schematic of T1 process is illustrated in Fig. 1.5. T2 events happen when one small bubble completely vanishes (cf. Fig.1.5).

1.4.2 Beyond the quasi-static limit

Subject to shear, a liquid foam may undergo structure changes. For instance, in a flowing foam when two bubbles come into contact with each other, the film between two bubbles can thin into a critical thickness in which van der Waals forces can trigger film instability and consequently rupture. Hence, two bubbles coalesce with each other and form a larger one. In addition, a fairly large bubble can break into smaller one in a flowing foam [27]. Golemanov et al. (2008) sheared a three dimensional foam in a parallel disk



Figure 1.5: Illustration of T1 and T2 events in dry foam.

geometry and observed bubble breakup above a critical shear stress that is two orders of magnitude smaller than that for breakage of a single bubble. T1 events do not happen just as a result of coarsening; also shearing of foam may trigger T1 events [17, 46]. Finally, in flow of a confined foam, bubbles with different sizes might interact with each other and induce crossstreamline migration and an effective diffusion. Herzhaft (2002) sheared a polydisperse foam in a similar geometry and observed segregation of bubbles across the gap according to the size; smaller bubbles were mostly found closer to the walls and bigger bubble at the middle of the gap. It is as yet not clear whether this might be due to breakup, coalescence or migration of bubbles or a combination thereof. Herzhaft (2002) also measured the shear stress in a parallel disk geometry and observed an overshoot in shear stress followed by reduction as a function of time in step strain tests. Interestingly, Golemanov et al. (2008) carried out similar experiments but observed that the shear stress increases as a function of time and attributed this to breakup of bubbles. Quilliet et al. (2005) observed migration of a bigger bubble in a sheared two-dimensional foam toward the periphery sides (i.e. towards the side walls). The morphological changes mentioned above affect the rheology and mechanical properties of the foam as a whole. Consequently, a fundamental understanding of the structural and rheological evolutions of foam is of scientific as well as practical importance [30].

1.5 Rheology of liquid foams

Although foams only contain fluids, they behave like viscoelastic solids or like non-Newtonian liquids, depending on applied stress, liquid volume fraction and the time scale set by the inverse of frequency in oscillatory experiments or the time elapsed since the application of a transient stress or strain. The rheological behavior of foams, concentrated emulsions, pastes and many other soft materials is strikingly similar. Slow relaxations, aging and jamming phenomena are found in all of these forms of soft condensed matter. Some might be due to generic mechanisms, acting on a mesosocopic length scale, while others might arise from the phyico-chemical composition of the materials. Subjected to small stresses, foam exhibits solid-like behavior. Beyond a yield stress, it behaves as a liquid-like material. The Herschel-Bulkley model is commonly used to describe foam rheology [62]. Thus, the geometrical, hydrodynamical and rheological properties of gas liquid foam can be tuned to make it a uniquely versatile multiphase mixture for a variety of process applications and product designs. It is therefore a material that is of broad interest to chemical engineers [72].

1.5.1 Experimental methods

Various experimental techniques are employed to characterize and analyze foam deformation and flow [18, 30, 53]. The macroscopic response to applied shear stress can be measured by conventional rheometers. For instance: Parallel plate, cone-plate and Couette cylinders have all been successfully used as shear geometries. However, several precautions must be taken to obtain physically interpretable rheological results. The surfaces of the confining walls must be roughened to avoid wall slip. Alternatively, one can use smooth surfaces and, in such cases, the foam-wall slip must be explicitly considered in data analysis [63]. In addition, measures must be taken to ensure foam stability during the experiment, with respect to liquid drainage, bubble coarsening and liquid evaporation at the contact with ambient atmosphere. Alternatively, one can study the coupling between foam aging (due to bubble coarsening or size-based segregation) and the rheological foam properties. In this case, the ageing process must be characterized for a foam sample, identical to that studied in the rheometer. For all these reasons, the rheological foam measurements are far from straightforward and the experimental protocols should be designed carefully, depending on the specific system and aim of the study.

Several methods have been used to characterize the bubble velocity profiles and structural rearrangement dynamics in flowing foams. Magnetic resonance imaging (MRI) detects the velocity distribution inside sheared foam, while diffusing wave spectroscopy (DWS) provides statistical information about the rate of bubble rearrangements in strained and in flowing foams [53, 60, 61]. Direct optical observations of bubble monolayers (2D foams) have provided rich information about the bubble shape and dynamics in flowing foams [39, 46, 70]. Direct observations of dynamics inside dry 3D foams have been carried out using optical tomography [69]. The experimental studies have clearly evidenced that the rheological response of foam involves processes in a wide range of length-scales. The deformation of individual bubbles creates the elastic stress of foams, while the yielding and plastic flow are the consequence of rearrangements in the bubble packing, and the viscous friction in the liquid films between neighbouring bubbles is a source of energy dissipation. At present, one of the most challenging and exciting research problems in foam rheology is to explain and predict the links between the macroscopically observed foam behavior and the microscopic processes that govern this behavior.

Our focus in this thesis will be on the mechanisms for structural evolution of foams in dynamic state and on the correlation between their rheology and structure.

Due to the opacity of the three-dimensional foam, it is hard to directly observe the changes in its structure during flow and correlate them to the rheological properties. To connect the rheology to the local behavior of bubbles, therefore, researchers have studied flow of two-dimensional foam which are monolayers of 3D bubbles. Experiments on two-dimensional foam flow have been performed in different geometries. For example some authors have confined foam bubbles in a Hele-Shaw cell in Couette geometry and rotated the inner disk while having the outer cylinder stationary [17]. They reported localized flow profiles in which the velocity profiles show fast decay away from the driving boundary. In the liquid-glass case, the foam is constricted from the top by a glass plate and at the bottom is in contact with a solution. For the third case, the bubble monolayer floats on the liquid and is exposed to air on top. This configuration is also known as the bubble raft. In Couette geometries, Dennin and co-workers have sheared bubble rafts with a fixed inner disk and a rotating outer cylinder [46]. More recently Katgert et al. (2010) studied both bubble rafts and two-dimensional foam with glassliquid boundary condition and showed that the normalized velocity profile is shear dependent in the presence of the upper wall and is independent of shear rate for a freely floating bubble raft [39]. They concluded that the boundary condition plays an important role on the localization of velocity profile.

All studies so far on shearing two-dimensional foams have been focused on low shear rates and the only topological changes observed is T1 events. For three dimensional foams Herzhaft (2002) and Golemanov et al. (2008) reported structural changes, but there is at present little understanding of the mechanism or mechanisms responsible for that structural evolution. In addition apparently there is contradiction in the evolution of shear stress in their measurements [27, 28]. Therefore, it is of high importance to measure the rheological properties of foam flow and see the effect of structural evolution on rheological properties. These are our motivations for carrying out a series of experiments to identify the mechanisms involved in structural evolution for the flow of two-dimensional foam. In the following we will be explaining the objectives of this work in detail.

1.6 Objectives and contributions of this research

We propose a series of experiments to identify and investigate the mechanisms responsible for structural evolution of freely floating 2D foams (bubble raft) in a Couette co-axial cylinder geometry undergoing shear. Details of the geometry will be given later. The objectives of the thesis are to answer these fundamental questions:

- (1) Will there be breakup or coalescence in a sheared two-dimensional monodisperse foam?
- (2) Does a bubble whose size is larger than the neighboring bubbles migrates laterally with respect to the flow stream-lines?
- (3) What if we introduce more than one large bubble in a sea of smaller bubbles? Would we see the size-based segregation in sample of bidisperse two-dimensional foam?
- (4) What is the contribution of non-Newtonian behavior of two-dimensional foam to the structural evolution?

As will be seen, these have been accomplished to a good degree in the research described in Chapters 3-6. The main contributions of the thesis can be summarized into four general items:

- We have discovered an anomalous coalescence mechanism, whereby bubbles coalesce for shear rates above a threshold, as opposed to below a critical rate, which is the normal scenario for bubble and drop coalescence. We also proposed an explanation for the anomalous coalescence.
- We reported for the first time, the lateral migration of a single large bubble in an otherwise monodisperse foam. This cross-streamline migration pushes the large bubble away from the walls. We developed a continuum model to account for the migration.
- We reported the size-based segregation in bidisperse and polydisperse two-dimensional foams and developed a model based on migration and shear induced diffusion to explain this process. We developed a migration-diffusion model that accurately predicts the size segregation.
- We studied the effect of non-Newtonian behavior of foam on bubble migration in a wide gap Couette device. We found that non-Newtonian rheology of foam changes the migration process and then we strived to explain it by separating the viscous and elastic contributions in migration process.

1.7 Thesis outline

Chapter 2 presents details of the experimental setup and research methodology. We describe a shear cell Couette device used for all the experiments to be presented in the thesis. Additionally, we explain the characterization of the material used in this research and at the end we present the visualizing technique that was utilized in this research.

Chapters 3-6 deal with the four research projects in turn. Chapter 3 presents detailed results of bubble-bubble coalescence in sheared twodimensional foam. We observed a threshold above which coalescence occurs. This threshold depends on the bubble size and liquid viscosity. Then, we offered several mechanisms to explain this anomalous coalescence. The most promising one is the model based on the centripetal force.

Chapter 4 presents new results on migration of a single large bubble in a sea of smaller bubbles. We report lateral migration of this single large bubble across the gap to a final equilibrium position which turned out to be the middle of the gap. The migration occurs above some thresholds for shear rate and bubble size ratio. We modify the Chan-Leal formula to explain the migration in foam and also used a force balance to explain the presence of thresholds for migration.

Chapter 5 presents experimental results on bidisperse and polydisperse two-dimensional foams. We report the size based segregation above thresholds for bubble size and shear rate, similar to migration study. Then, we develop a model to account for shear induce diffusion as well as lateral migration.

Chapter 6 presents an extension to chapter 4 in which we have widen the gap of the Couette cell device. We showed that this seemingly naive change in geometry elicits the non-Newtonian behavior of foam. Again, migration of a single bubble in an otherwise monodisperse foam was investigated, but this time bubble migrates to a final equilibrium position which is not at the middle of the gap anymore. Modification of Chan-Leal formula no longer predicts the experimental observations. Hence, the viscoelastic nature of foam comes to play. We rationalize the bubble migration experiments in foam by studying the migration in shear thinning and Boger fluids which mimic the foam behavior.

Chapter 7 summarizes the key results of the thesis, outlines the limitations of the current work, and makes recommendations for future work.

Chapter 2

Experimental setup and methodology of research

2.1 Materials

Since the experiments are performed on the two-dimensional liquid foam, the materials that we need to make the foam include: distilled water, glycerol (Fisher Scientific), dish washing liquid (Unilever, Sunlight) and nitrogen, which will be provided by Praxair company in compressed steel cylinders. In addition to that for other experiments we have used xanthan gum (West Point Naturals) and Polyacrylamide (Sigma-Aldrich, Mw = 5,000,000-6,000,000).

2.2 Material characterization

Prior to the actual experiments on the foam flow, we have performed some experiments to characterize the materials that are present in the experiments.

- Surface tension: Surface tension was measured by a tensiometer (Cole-Parmer, Surface Tensiomat 21 WU-59951-14) which is a Du Noy Ring type Tensiometer. We measured surface tension for different liquids, including the pure liquids and soap solution at room temperature. This type of tensiometer uses a platinum ring which is fully submerged in a liquid. As the ring is pulled out of the liquid, the tension required is precisely measured in order to determine the surface tension of the liquid.
- **Density**: Density of different fluids is measured by using a Density Meter (Anton- Paar DMA 35N) at room temperature.
- **Rheology**: Rheological properties such as shear viscosity and shear stress of all pure materials and soap solutions as well as two-dimensional foam are measured by using a rheometer (Malvern, Kinexus) and MCR (502) with a co-axial cylinder and cone-plate geometry.



Figure 2.1: Schematic of the shear cell (not to scale). (b) A top view snapshot of 2D foam at rest with bubble radius of 500 μm .

2.3 Shear cell device and foam making procedure

The migration experiments are carried out in a modified Couette cell device. This cell consists of two cylinders: a stationary outer cylinder with the inner radius of $R_0 = 10 \ cm$ and a rotating sharp-edged inner disk with two different radii, $R_i = 9.3 \ and \ 8.1 \ cm$ (Fig. 1a). The static liquid level is flush with the top surface of the inner and outer cylinders such that the interface is pinned at the sharp corners. Furthermore, triangular teeth are machined onto the solid surfaces to prevent slippage of first row of bubbles. Bubbles are produced by blowing nitrogen through an immersed capillary tube in a soapy solution using a pneumatic PicoPump (WPI, model PV-820). This method allows us to make an extremely uniform bubble size that can be fine-tuned by the nitrogen pressure. Moreover, the inner cylinder is attached to a servomotor that can be rotated from 0.1rpm to 100rpm using a motor controller.



Figure 2.2: (a) A top view snapshot of 2D foam at rest with bubble radius of $\approx 540 \ \mu m$. (b) Typical velocity field of foam obtained by PIV at $\Omega = 5 \ rpm$.

2.4 Imaging techniques and bubble size measurement

Multiple cameras have been used for the experiments. We used a high-speed camera (Megaspeed, MS 70K) to capture the evolving foam structure. The frame rate ranges from 25 fps to 20,000 fps. In addition to that, we used two other high-resolution cameras (Watec model 902B) to directly observe the structure of the foam and to measure the bubble velocity profile across the gap by using Particle Image Velocimetry (PIV). For PIV we used an open source Graphical User Interface (GUI) code in MATLAB developed by William Thielicke to track the position of bubbles in consecutive exposures [74]. By measuring the displacement of bubbles between consecutive images we can calculate the foam velocity profile across the gap. Fig. 2.2 shows top view snapshots of foam at rest (Fig. 2.2 (a)) and typical velocity vectors obtained PIV method for sheared foam (Fig. 2.2 (b)). We also measured the bubble size and bubble size distribution using microstructural measurement software developed by Nahamin Pardazan Asia Co. [80]

Chapter 3

Coalescence of bubbles in sheared two-dimensional monodisperse foam

3.1 Introduction

As discussed in chapter 1, foams are fragile soft matter, with a microstructure which is thermodynamically and mechanically unstable [2, 6, 68, 76]. For three-dimensional foams there have been number of studies which investigated the structural evolution in flowing foam. For instance, Golemanov et al. [27] observed breakup of bubbles sheared between parallel disks. On the other hand, in a similar geometry, Herzhaft [28] reported size-based segregation of bubbles in a polydisperse foam in which smaller bubbles found to be close to the top and bottom disks while larger ones mainly accumulated in the middle. Conspicuously missing, however, is any report of shear induced coalescence, a common occurrence in sheared emulsions [58]. This has motivated us to study the possibility of bubble-bubble coalescence in a sheared foam. The most important difficulty with understanding the structure-flow coupling in a 3D foam emanates from its opacity; Therefore, recently researchers have mainly focused on experimentation with 2D foams, i.e. monolayers of 3D bubbles. This way, they can easily see and correlate the microstructure to flow properties. The only structural changes observed so far are T1 events [46]. Our main goal in this study is to investigate the structural changes in 2D foams under more vigorous shearing. The main finding is a new type of bubble coalescence unexpected at the start. We consider several models for the anomalous coalescence, the most promising one is the one based on inertia of the fluid which can explain most of experimental observations.

3.2 Results

3.2.1 Critical rotational velocity for coalescence

In this work we found that there is a threshold for rotational velocity Ω_c above which large bubbles start to appear quickly after the start of shearing



Figure 3.1: (a) A foam with bubble radius $R = 500 \ \mu \text{m}$ in liquid I shows no sign of coalescence when sheared at 60 rpm. (b) Large bubbles appear after about 20 seconds of shearing at 75 rpm.

(Fig. 3.1). At shear rates below Ω_c , no large bubbles appear during the long period of experiments and the foam morphology remains the same as the beginning($\sim 30 \text{ min}$). The threshold is observed for all bubble sizes, liquid compositions and surfactant concentrations that we tested.

Our optical setup offers a 2 cm \times 2 cm viewing window that is fixed in space. Thus, we capture only a small portion of the circular trajectory of the bubbles. The coalescence takes place very quickly after shearing above Ω_c . Therefore, we cannot capture the actual process of the coalescence as Ritacco et al. [68] did for bursting of bubbles in static bubble raft. Coalescence is thus inferred from the appearance of large bubbles.

This surprising result is not in line with the conventional wisdom that coalescence happens for gentler collisions, with an *upper* bound on the shear rate and a corresponding maximum capillary number [e.g. 10, 12, 19, 75, 81]. The coalescence between two freely suspended bubbles or drops is determined by the competition between two time scales, the interaction time t_i and the drainage time t_d . The former is the time that the two bubbles spend to interact with each other, and scales with the inverse shear rate, while the latter is the time required for the liquid film between them to drain to a critical thickness such that van der Waals forces can trigger

Solution	Glycerin	С	$\mu \text{ (mPa·s)}$	$\sigma (mN/m)$
Ι	10 wt.%	5 wt.%	1.0 ± 0.1	27.0 ± 1.0
II	30 wt.%	5 wt.%	1.8 ± 0.2	27.0 ± 1.0
III	50 wt.%	5 wt.%	4.2 ± 0.4	27.0 ± 1.0

Table 3.1: Composition and properties of the solutions.

the rupture of film [10]. The requirement of $t_i \gtrsim t_d$ for coalescence leads to an upper critical capillary number. Such a criterion has been verified by extensive studies that examined various parameters in the process, including drop size, viscosity of the fluids, lateral offset of the colliding drops, and surfactant concentration [e.g. 32, 33]. This apparently does not apply in our case.

3.2.2 Effect of bubble size and liquid viscosity

We have examined the effects of the bubble size R and liquid viscosity μ on coalescence process. Fig. 3.2 shows that the critical angular velocity Ω_c increases with both R and μ . This is again surprising: it implies that the anomalous coalescence cannot be analyzed in the conventional framework of a capillary number, i.e. in terms of viscous forces competing with surface tension. There must be a new mechanism at play that was absent in the convention scenario of collision and coalescence.

If we draw straight lines through the data points in this log-log plot, their slopes give the scaling $\Omega_c \propto R^{0.27\pm0.02}$. The dependence of Ω_c on the liquid viscosity μ is rather weak: $\Omega_c \propto \mu^{0.1}$.

3.2.3 Interfacial shape and bubble distribution

We have also recorded the shape of the foam-air interface and spatial redistribution of the bubbles under shear. These may offer potential clues to the cause of the anomalous coalescence.

Intuitively one may expect the centripetal force to deform the interface on the rotating liquid. This is not the case; the interface exhibits no observable variation in its elevation across the gap even at the highest Ω tested. This is largely due to the effect of pinning of the interface at the solid walls. In the experiment, we fill the gap between the cylinders such that the static liquid surface is flush with the tops of the cylinders. Once shearing starts, the free surface is subject to the centripetal force as well as anchoring on the sharp edges of the inner and outer walls. Using the velocity profiles of Fig.3.3, we have computed the shape of the interface, shown in Fig. 3.4 for a rotational speed of 60 rpm. Thus, the anchoring of the surface limits its undulation to negligible amounts (< 0.2 mm; one order of magnitude smaller than without anchoring). Furthermore, the bubbles are held mostly



Figure 3.2: Critical rotation speed Ω_c as a function of bubble radius R for liquids I, II and III (see Table 3.1).



Figure 3.3: Bubble velocity profiles at three rotation speeds with liquid I. Bubble size $R = 250 \ \mu\text{m}$. The line represents the analytical solution for a Newtonian fluid.



Figure 3.4: Shape of the free surface for liquid sheared at $\Omega = 60$ rpm. The radial distance is measured from the axis of rotation and the inner and outer boundaries are at 93 mm and 100 mm, respectively.

underwater by surface tension [54].

At relatively low rotation speed, the bubbles slide past each other in rows. At higher Ω , however, there appears to be radial motion of the bubbles that disrupts the layers. As a result, bubbles tend to be more tightly packed in the inner half of the gap than in the outer half. In fact, voids of clear liquid start to appear in the outer region (Fig. 3.5), which quickly disappear after the shearing stops. The most plausible cause of this spatial inhomogeneity is the centripetal force exerted by the rotating liquid on the bubbles. This may be the cause of this anomalous coalescence as explained below.

3.3 Potential mechanisms for anomalous coalescence

One should not that there are some differences between our experiments and previously reported drop- or bubble-coalescence experiments. In shearinduced drop collision, the shear brings into contact two freely suspended drops that would otherwise not interact with each other at all. In our bubble raft, on the other hand, bubbles are in close contact with each other even without shear. Why should the static bubbles be immune to coalescence while the sheared ones are not? Moreover, our bubbles are covered by surfactants, and there is also ample supply of it in the surrounding liquid. Finally, our coalescence occurs at relatively high flow rates, much higher



Figure 3.5: Bubble distribution in a foam 10 min after shearing at $\Omega = 60$ rpm. $R = 500 \ \mu$ m.

than typical of drop-coalescence experiments [83]. In the following, we will explore these differences for clues to the anomalous coalescence.

3.3.1 Shear precludes surfactant-stabilized films

The first idea is to investigate difference between the stability of static foam and the coalescence in a sheared one. Static foams are stabilized by surfactants because the latter form regular structures in liquid films that are sufficiently thin [64]. If the bulk surfactant concentration is below CMC, bilayers of surfactants form the so-called "black films" [14]. At higher concentrations, micelles arrange themselves into a more or less regular colloidal structure in the liquid film, producing thick stable films [64]. In either case, the surfactant structure contributes a disjoining pressure that prevents liquid drainage and stabilizes the static foam. Conceivably, vigorous shearing may disrupt such surfactant structures or prevent them from forming in the first place. This could be a mechanism for the observed coalescence.

In a recent study, Denkov et al. [20] demonstrated how the black film may cause jamming in flowing foams. In essence, they assume that for low enough shear rates, there is enough time for the film between neighboring bubbles to thin down to a critical thickness where attractive forces act to produce black films. Then the bubbles are locked into a rigid structure that resists the shearing, and the foam is jammed. In our experiment, the surfactant concentration is above CMC and the stable structure should be the thick stable film instead of the black film [64].

At low Ω , we observe nonhomogeneous shearing with large domains of jammed bubbles. Around $\Omega = 3$ rpm, all such domains unjam and the bubble raft starts to shear more or less uniformly. We thus take this to be the threshold for the destruction of the thick stable films. However, larger bubbles only start to appear at a much higher rotational speed of $\Omega \sim$

60 rpm. Therefore, the unjamming cannot be the cause of the anomalous coalescence, which requires much more vigorous shearing.

3.3.2 Surface remobilization due to surfactant transport

The second mechanism which might be responsible for coalescence in foam is surfactant transport. Drainage in liquid films carries surfactants along the interface, and creates a gradient in surfactant concentration along the interface of bubbles. This in turn produces a tangential Marangoni stress that resists the interfacial flow. Conceivably, sufficiently strong shearing may produce a viscous stress τ_v that overpowers the Marangoni stress τ_M , thereby remobilizing the bubble surfaces. Once bubble surface is mobilized the film drainage will be facilitated and so will coalescence. This suggests using the Marangoni number $Ma = \tau_M/\tau_v \sim 1$ as a criterion for the observed anomalous coalescence. In studying pairwise collision of surfactant-covered drops, Yoon et al. [83] used this argument to rationalize the appearance of a "transition capillary number" for lower bulk surfactant concentrations such that coalescence occurs *above* it but not below. This seems to be consistent with our anomalous coalescence.

Therefore, we will study the antagonism between Marangoni stress and viscous stress as a potential explanation for the anomalous coalescence observed in our experiment. For soluble surfactants, the surface concentration is determined by two steps: bulk diffusion of surfactants toward the interface and adsorption onto the interface [49]. For our commercial detergent, it is not possible to estimate the relative rates of these two steps. We will examine the cases of either one being the limiting step by adapting the classical analysis of Levich on falling drops [49]. In our problem, the liquid flow outside the bubbles is due to shear instead of sedimentation. Thus, we need to replace the characteristic liquid velocity in Levich's calculations by $\dot{\gamma}R$, $\dot{\gamma}$ being the local shear rate.

If adsorption is the limiting step that dictates the surfactant distribution Γ on the bubble surface, one can estimate the surface concentration gradient as [49]:

$$|\nabla \Gamma| \approx \frac{\Gamma_0 \, \dot{\gamma}}{\alpha \, R},\tag{3.1}$$

where Γ_0 is the equilibrium concentration and α is the coefficient of adsorption. This implies that the Marangoni stress

$$\tau_M = |\nabla\sigma| = \left(\frac{\partial\sigma}{\partial\Gamma}\right)_{\Gamma_0} |\nabla\Gamma| \tag{3.2}$$

is proportional to the shear rate. Since the shear stress τ_v on the surface is also proportional to $\dot{\gamma}$, the ratio τ_M/τ_v will be independent of the shear rate. This cannot explain the fact that coalescence happens above a threshold rotational speed. When the bulk diffusion determines the surfactant distribution on the bubble surface, Levich [49] estimated $|\nabla\Gamma|$ and hence $|\nabla\sigma|$ based on a boundary layer thickness $\delta \sim (DR/\dot{\gamma})^{1/3}$, D being the bulk diffusivity:

$$\tau_M = |\nabla\sigma| \approx \frac{\Gamma_0 \dot{\gamma} \delta}{DR} \left(\frac{\partial\sigma}{\partial c}\right), \qquad (3.3)$$

where c is the bulk concentration of the surfactant, and $\partial \sigma / \partial c = \Gamma_0 R_g T / c$ by virtue of the Gibbs equation, R_g and T being the gas constant and absolute temperature. Now the stress ratio can be written as

$$\frac{\tau_M}{\tau_v} \approx \frac{\Gamma_0^2 R_g T}{c} \frac{\delta}{\mu D R}.$$
(3.4)

From the Stokes-Einstein relationship, the surfactant diffusivity D is inversely proportional to the liquid viscosity μ : $D = k_B T/(6\pi\mu r_s)$, r_s being the characteristic size of the surfactants and k_B the Boltzmann constant. Plugging this and the estimation of δ into the above equation, we obtain

$$\frac{\tau_M}{\tau_v} \approx C \,(\mu \dot{\gamma} R^2)^{-1/3},\tag{3.5}$$

where C contains factors including T and c, and is a constant in our experiment. The prediction that τ_M/τ_v decreases with $\dot{\gamma}$ allows the possibility that the Marangoni stress be overpowered by the viscous shear stress at sufficiently high $\dot{\gamma}$, which would be consistent with the proposed mechanism of bubble-surface remobilization. However, the prediction of a critical shear rate that scales with μ^{-1} and R^{-2} contradicts the observations in Fig. 3.2.

In view of the above analysis, we are driven to the conclusion that the remobilization of bubble surface by shear stress overcoming Marangoni stress cannot be the cause of the anomalous coalescence.

3.3.3 Bubble compression due to inertia

The photo in Fig. 3.5 indicates a tendency for the bubbles to be pushed radially inward. The only plausible agent for such an effect is the centripetal force of the rotating liquid. As the spinning liquid generates an inward pressure gradient, the bubbles, having a much lower density than the liquid, are pushed inward towards the inner cylinder. Thanks to pinning on the walls, the liquid surface rises little (Fig. 3.4). The radial pressure gradient is thus maintained not by hydrostatic head but by surface tension in the liquid meniscus. Conceivably the squeezing between bubbles accelerates the drainage in the liquid film. If the film drains down to a critical thickness within the interaction time between two bubbles, coalescence would occur [10]. Thus, one may be able to adapt ideas from conventional drop-drop collision to explain the anomalous coalescence. In the following we test this mechanism through a scaling model.

For a pair of bubbles pushed into each other by a constant force F, we may estimate the drainage time from an initial film thickness h_0 to the final critical one h_c using the rigid parallel disk model [10, 52]:

$$t_d = \frac{3\pi\mu a^4}{4F} \left(\frac{1}{h_c^2} - \frac{1}{h_0^2}\right),\tag{3.6}$$

where a is the radius of the liquid film. In our geometry, the radial pressure gradient due to the spinning liquid is $dp/dr = \rho u^2/r$, ρ being the liquid density and u the tangential velocity of the liquid at distance r. This exerts a force $(dp/dr) \cdot 2R \cdot \pi R^2$ on each bubble. Since the bubbles are in close contact with each other, they transmit the centripetal force onto their inner neighbors in a sort of force chain, resulting in the largest cumulative force on the innermost layer of bubbles:

$$F = 2\pi\rho R^3 \sum_{i=1}^{N-1} \frac{u^2}{r},$$
(3.7)

with the summation over the outer layers of bubbles. In comparison with F, the squeezing force $\pi a^2(\sigma/R)$ due to capillary pressure is at least an order of magnitude smaller, and has thus been neglected.

Chesters and Bazhlekov [11] have proposed an empirical relation for the critical film thickness h_c for rupture due to van der Waals force:

$$h_c = \frac{2}{3} \left(\frac{A}{4\pi\sigma}\right)^{0.3} (aR)^{0.2}, \qquad (3.8)$$

A being the Hamaker constant taken here to be $A = 3 \times 10^{-19}$ J [35]. We need now to estimate a. For pairwise collisions in a shear flow, the classical theory gives $a/R \sim Ca^{1/2}$ [10]. We have measured a directly by using ImageJ [67], and found it relatively insensitive to shear. In the static foam, $a \approx 0.17R$, in close agreement with previous computations [16]. With shearing, a tends to increase with Ω but quickly saturates to an average value $a \approx 0.2R$ at about 25 rpm. Apparently the close packing constrains the bubble movement and diminishes the role of shearing. Measuring a among hundreds of pairs of bubbles reveals moderate variations in any given foam, and Fig. 3.6 shows a typical distribution of a in a sheared foam. Since smaller a gives faster film drainage, and we are concerned with the onset of coalescence, we use the smallest a = 0.14R. Inserting this value along with Eqs. (3.7) and (3.8) into Eq. (3.6), the ratio between drainage and interaction times is:

$$\tau = \frac{t_d}{t_i} = 1.64 \times 10^{-3} \left(\frac{\pi\sigma}{A}\right)^{0.6} \frac{\mu R^{0.2} \dot{\gamma}}{\rho \sum_{i=1}^{N-1} \frac{u^2}{r}}.$$
(3.9)

where we have neglected h_0^{-2} relative to h_c^{-2} , and taken the interaction time between neighboring rows of bubbles to be $t_i \approx \dot{\gamma}^{-1}$ as in previous analysis


Figure 3.6: Distribution of the film radius a in a sheared bubble raft with bubble size of $R = 500 \ \mu \text{m}$ undergoing rotational speed of $\Omega = 75 \text{ rpm}$. The curve shows a fitted normal distribution.



Figure 3.7: The critical condition for coalescence corresponds to $\tau = 8$ for solution I, τ being computed for the innermost layer of bubbles using the measured bubble velocity profile. For solutions II and III, the critical τ values are 12.5 and 28.5, respectively.

[10]. We argue that $\tau \leq O(1)$ should give the critical condition for the anomalous coalescence observed here.

The validity of the scaling theory can now be tested against the key experimental observations. First, note that $\dot{\gamma}$ and u are both proportional to Ω . Thus $\tau \sim \Omega^{-1}$ and $\tau < 1$ does yield a minimum critical rotational speed as observed. Quantitatively, however, the critical condition corresponds to $\tau = 8$, 12.5 and 28.5 for solutions I, II and III, respectively (Fig. 3.7). These numbers are one order of magnitude too large. Second, the τ criterion predicts a scaling for the critical rotational speed $\Omega_c \sim R^{0.2}$, in reasonable agreement with the power-law scaling observed in Fig. 3.2. Third, it also predicts Ω_c to increase linearly with the liquid viscosity μ . While the trend is correct, the experimentally observed dependence on μ is much weaker: $\Omega_c \sim \mu^{0.1}$ (cf. Fig. 3.2). Finally, the large bubbles appear more often in the inner part of the gap than the outer. Given that the smallest a can be anywhere in a particular experiment, this provides indirect support for the accumulation of the inward force in Eq. (3.7).

Thus, the inertia-based mechanism explains the qualitative trends observed. But quantitatively it overestimates the drainage time as well as the effect of liquid viscosity. The latter recalls the study of Yoon et al. [82] on freely suspended droplets, where the viscosity effect is also weaker than expected. In our case, the numerical discrepancies have many potential causes. For example, the Hamaker constant [35] is not known for the fluids used here, and possibly the bubble surface may develop dimples during thinning [75] that would compromise the calculation above. Since our bulk surfactant concentration is 100 times CMC, the abundance of surfactants may introduce additional effects. Rapid adsorption onto the bubble surface may partially mitigate the Marangoni stress and locally remobilize the surfaces [64]. Though this has been dismissed as a critical condition for the anomalous coalescence, it might explain the fact that the drainage rate is underestimated in our model, producing too large a critical τ value. Moreover, the later stage of drainage is probably influenced by the presence of micelles, which may form layers that hinder film thinning below $h \sim 100 \text{ nm}$ [64]. This non-viscous effect may reduce the overall dependence on μ . Unfortunately, not knowing the chemical properties of the surfactant mixture in the detergent, it is difficult to formulate these ideas quantitatively.

3.4 Conclusion

In this chapter we reported an anomalous type of bubble coalescence in a monolayer sheared in a Couette device, which occurs above a critical rotational speed Ω_c . This contrasts the conventional wisdom about bubble and drop coalescence that it occurs below a critical capillary number. Our coalescence cannot be characterized by a critical capillary number; the critical Ω_c increases with bubble size and the viscosity of the suspending liquid. To rationalize the experimental observations, we have considered three potential mechanisms for the coalescence: shear preventing the formation of surfactant-stabilized films between bubbles, shear stress overcoming Marangoni stress to remobilized the bubble surface, and centripetal force pressing the bubbles radially inward into each other. None of these accounts quantitatively for all the experimental results.

The third is the most promising. According to this model, the anomalousness of the scenario arises from two factors: the film drainage is driven by a centripetal force instead of a viscous one, and the bubble deformation is determined by geometric constraints rather than shearing. The apparent reversal in the coalescence criterion, from the conventional maximum capillary number to a minimum shear rate, is similar in spirit to that demonstrated recently by Ramachandran and Leal [66] for collision between vesicles. Though clearly not a complete theory for the anomalous coalescence, the inertia-based model captures the qualitative trends of the experiment, and may serve as a starting point for further investigations.

Chapter 4

Lateral migration of a single large bubble in monodisperse two-dimensional foam

4.1 Introduction

Foams rheology and hydrodynamics are intimately coupled to its microstructure, i.e. the shape and spatial organization of the bubbles [30, 78]. A particularly intriguing phenomenon is size-based segregation of bubbles in a polydisperse foam [28]. After shearing between rotating parallel plates, smaller bubbles appear predominantly near the top and bottom plates while the larger ones are in the middle. The cause is unclear, but one possibility is that the bubbles have migrated across streamlines based on their size. In a more recent experiment on a two-dimensional (2D) foam under oscillatory shear [65], a bubble larger than its monodisperse neighbors migrates toward one of the four borders confining the foam. This seems to contradict the observations of Herzhaft on size-based segregation of bubbles [28]. More curiously, the migration does not distinguish between the flow direction and the direction of the velocity gradient. These two studies hint at some rule governing lateral migration of bubbles in sheared foam, but little is known at present. In contrast, lateral migration of particles and droplets suspended in a liquid medium has been extensively studied in the past [e.g. 7, 8, 34, 47]. A solid spherical particle in a Stokes flow cannot migrate because the linear system is time-reversible. A droplet deforms under shear, and this introduces a nonlinearity into the problem and makes lateral migration possible. It has been shown that in low-Reynolds-number Couette flows, droplets move away from the walls toward the center of the gap [8, 31, 38]. This is commonly interpreted as a wall repulsion; the rigid wall produces an asymmetry in the velocity and pressure fields around the drop. Hence arises the lateral migration force. Naturally one wonders if the same repulsion operates in sheared foam. This chapter describes an experimental study of lateral migration of bubbles in a 2D foam sheared steadily in a narrow-gap Couette device. Into a monodisperse bubble raft we introduce a single bubble of different size and investigate its migration. By correlating the migration speed with the shear rate and the bubble size ratio, we propose a hydrodynamic explanation for





Figure 4.1: Migration trajectories of a single bubble (R = 0.7 mm) at $\Omega = 7 \text{ rpm}$. The curves represent the Chan-Leal formula (Eq. 4.1). The top inset illustrates the migration schematically (not to scale) and the bottom one depicts the liquid meniscus above the bubble calculated from the model of Ref. [54].

the migration based on bubble deformation.

4.2 Results

4.2.1 Migration of bubble in a Newtonian fluid

As a baseline, we first study the migration of a single bubble floating on the free surface. It migrates to the center of the gap from all initial positions. Typical trajectories are shown in Fig. 4.1. The dimensionless drop position s is scaled by the gap width d, with s = 0 at the inner cylinder and 1 at the outer cylinder. The symmetry between inward and outward trajectories confirms the uniformity of the shear rate across the gap. This migration is reminiscent of that of neutrally buoyant droplets suspended in a liquid medium [8, 31]. Thus we have compared the measured trajectories with those predicted by the theory of Chan and Leal [8]. Chan and Leal [8]

considered the migration of a Newtonian drop in a Newtonian matrix sheared in a Couette device, under the condition of vanishing capillary number and small drop deformation. For a bubble of radius R in a matrix of viscosity η , the dimensional migration velocity can be written as:

$$v_m(S) = \frac{\sigma}{\eta} Ca^2 (\frac{R_i}{R_o})^4 \{ \frac{81}{560} \frac{R^2}{d^2} [1 + \frac{R_o^2}{(R_i + Sd)^2}]^2 f(S) - \frac{1}{7} \frac{R_o^4 R}{(R_i + Sd)^5} \}, \quad (4.1)$$

where $Ca = \eta \dot{\gamma} R / \sigma$ is defined using the shear rate at the inner cylinder. $f(S) = S^{-1} - (1 - S)^{-2} + 2 - 4S$, and we have put the bubble viscosity to zero. The first term in the bracket represents wall repulsion that pushes the bubble to the center of the gap (S = 0.5), while the second term is due to the curvature of the streamlines and drives the bubble toward the inner cylinder. Thus, the Chan-Leal formula predicts an equilibrium position between the center and the inner cylinder. Note that the prerequisites for the perturbation theory, $Re \approx 0$, $Ca \ll 1$ and $R \ll d$, are all satisfied by the experiment. With Re, Ca, R and d being Reynolds number, capillary number, bubble radius and gap size respectively. Integrating the above using the experimental parameters produces the trajectories of Fig. 4.1. The agreement between the measured and predicted trajectories is very close. The formula was derived for a neutrally buoyant drop inside a 3D fluid while our bubble "floats" on the liquid surface. In reality, surface tension keeps 99% of the bubble volume below the undisturbed free surface, which is consistent with theoretical calculations [54] (Fig. 4.1 inset). The viscous friction in the thin meniscus atop the bubble may be larger than in a fully 3D geometry. But apparently the left-right asymmetry dominates and the vertical dimension seems to matter little. Thus, the Chan-Leal formula predicts the migration in our geometry with no fitting parameter.

4.2.2 Migration in two-dimensional monodisperse foam

The main result of the experiment is the migration of a larger bubble of radius R in an otherwise monodisperse bubble raft of radius r. Generally the large bubble migrates toward the center of the gap, and the migration speed depends on the size ratio $\kappa = R/r$ as well as rotation rate Ω . Figure 4.2 shows migration trajectories for several κ and Ω values. During the migration, the large bubble shifts from one row of bubbles to the next, spending a finite time in each. This is indicated by the horizontal bars on some trajectories, forming a staircase pattern. For clarity, the bars are omitted on the other trajectories with only data points plotted at the center of each step.

The following observations can be made. (i) There are a threshold κ_0 for a fixed Ω and a threshold Ω_0 for a fixed κ , below which no migration occurs. For the conditions of Fig. 4.2(a), κ_0 lies between 1.43 and 1.54. In particular, a bubble *smaller* than its neighbors, i.e. with $\kappa < 1$, does



Figure 4.2: Bubble migration in a 2D foam. (a) Effect of the bubble size ratio κ at $\Omega = 4$ rpm. The bubble radii are (in mm): (r, R) = (0.35, 0.5) for $\kappa = 1.43$; (0.39, 0.6) for $\kappa = 1.54$; (0.435, 0.7) for $\kappa = 1.61$ and (0.35, 1) for $\kappa = 2.86$. The solid and dashed curves are predictions of Eq. (4.3) for $\kappa = 1.61$ and 2.86. (b) Effect of the rotational rate Ω for fixed bubble sizes (r, R) = (0.35, 0.7) mm. The curves are predictions of Eq. (4.3) for $\Omega = 3$ and 7 rpm.



Figure 4.3: Bubble deformation in different environments at $\Omega = 7$ rpm. The large bubble (R = 0.7 mm) in (a) deforms much more in a foam of smaller bubbles (r = 0.35 mm) than alone in (b). (c) A smaller bubble (R = 0.4 mm) is shielded by its neighbors (r = 0.58 mm).

not migrate at all. In Fig. 4.2(b), Ω_0 is between 2 and 2.5 rpm. (ii) For sufficiently large Ω and κ , a large bubble migrates all the way to the center (s = 0.5). Below these, the bubble may migrate to an intermediate position between the wall and the center. (iii) The migration speed increases with κ and Ω . (iv) The migration is much faster than if a bubble of radius Rmigrates on a free surface, without the bubble raft. This can be seen by comparing Fig. 4.2 with Fig. 4.1; the migration time differs by a factor of $O(10^2)$.

4.3 A hydrodynamic model to explain the migration in foam

4.3.1 Model development

All the above observations can be explained by a model based on the deformation of the migrating bubble. Chan and Leal [8] showed that the wall repulsion stems from the left-right asymmetry in the flow around the bubble and the concomitant asymmetric bubble shape. In our experiment, a larger bubble protrudes outside its own row and forces the surrounding bubbles to rearrange as they pass around it (Fig. 4.3a). Compared to fluid particles in a continuum, the surrounding bubbles have a finite radius r and a capillary pressure inside, and thus are much harder to displace and deform. They continuously rub and bump into the sides of the large bubble, imparting a force F_b on it. This force is the counterpart of the liquid pressure and viscous force in the single-bubble scenario, but is much larger. A visible consequence of F_b is the pronounced deformation of the large bubble, much more than a single bubble of the same size subject to the same shear rate (Fig. 4.3b). A less visible one, we surmise, is a strong wall repulsion arising



Figure 4.4: Deformation parameter of a larger bubble in a 2D foam as a function of the bubble size ratio κ . The error bars indicate the variation among 7 shear rates tested, and the curve is a quadratic fit to the data.

from the asymmetry in F_b from the two sides.

This idea can be made more precise by plotting the bubble deformation as a function of the size ratio κ (Fig. 4.4). We define a bubble deformation parameter D = (l - a)/(l + a), l and a being the long and short axes of the roughly elliptical deformed bubble. According to Taylor's celebrated formula [73], a single bubble of negligible internal viscosity in a sheared fluid should have D = Ca. In the bubble raft, we represent the data by $D/Ca = g(\kappa) = 2.5\kappa^2 - 7\kappa + 11$. Now we equate the larger deformation in a bubble raft to that of a single bubble at a higher "effective capillary number" Ca_e :

$$Ca_e = D = Ca \cdot g(\kappa). \tag{4.2}$$

Plugging this into the Chan-Leal formula (Eq. 4.1) gives us a modified Chan-Leal formula

$$v_m(s,\kappa) = \frac{81}{140} \frac{R^2}{d^2} \frac{\sigma C a_e^2}{\mu} f(s) g^2(\kappa).$$
(4.3)

After time integration, this formula predicts well all the migration trajectories recorded in our 2D foam, over the entire range of r, R and Ω values. For clarity, only a few representative curves are plotted in Fig. 4.2. Note that the O(10) deformation enhancement in Fig. 4.4 translates to the $O(10^2)$ increase in the migration velocity. The success of Eq. (4.3) confirms our hypothesis in the preceding paragraph. As a corollary, a bubble of the same size as its neighbors or smaller ($\kappa \leq 1$) does not migrate because it does not jut out of its own row (Fig. 4.3c). Thus, it is not subject to the "bumping force" F_b .

4.3.2 Thresholds for migration

Finally, we examine the thresholds κ_0 and Ω_0 for lateral migration. When a monodisperse 2D foam is sheared, the bubbles typically move in streamwise rows past one another. For a larger bubble (radius R) to migrate laterally, it must squeeze into the next row of bubbles (radius r). The wall repulsion force F_w driving the migration, therefore, must exceed a threshold in order to deform the bubbles of the next row to create the gap. Because these bubbles in turn interact with multiple moving and changing neighbors on the other side, it is difficult to posit a precise force balance from which to calculate the threshold. As an estimation, we take the resistance to migration to be on the same order of magnitude as the *capillary force* between bubbles in the row: $F_c = (\sigma/r) \cdot \pi a^2$, where a is the radius of the thin film between neighboring bubbles in a 2D foam. For the foam quality used here, a shows a normal distribution among the bubble pairs, with a mean of a = 0.2r [57], which will be used below. On the other hand, the wall repulsion can be estimated from the Stokes formula using the migration velocity of Eq. (4.3): $F_w = 6\pi\mu v_m R$. We use the Stokes formula as opposed to the Hadamard formula because the bubble surface is immobilized by the high surfactant concentration [57]. Now the ratio between these two forces is:

$$\Phi = \frac{F_w}{F_c} = \frac{243}{70} \frac{R^3 r}{a^2 d^2} C a^2 f g^2.$$
(4.4)

Note that f(s) gives the wall repulsion at position S. In particular, using the largest F_w , for the first row next to the wall, gives us the ratio Φ_1 . We argue that a Φ_1 value of O(1) gives the threshold for lateral migration of the larger bubble.

Figure 4.5 plots Φ_1 for all the κ and Ω values tested in our experiments. The experimental conditions giving rise to lateral migration are indicated by filled and half-filled symbols, the latter for *partial migration* to positions between the wall and the center. The non-migrating conditions are shown by hollow symbols. These two groups are almost perfectly separated by $\Phi_1 =$ 0.4, thus validating Eq. (4.4) as an approximation for the threshold. For lower Ω , three data points fall on the wrong side of the line; the experiment may have been more susceptible to external disturbances in these cases.



Figure 4.5: Threshold for bubble migration. Φ_1 , computed from Eq. (4.4) for the first row of the bubbles next to the wall, is plotted as functions of Ω for various κ values. Hollow, half-filled and filled symbols indicate no migration, partial migration and complete migration to the center. The dashed line is $\Phi_1 = 0.4$.

Note that the thresholds reflect the graininess of the bubble raft, and have no counterpart in the Chan-Leal theory.

4.4 Conclusion

In this chapter, we studied lateral migration of a single bubble whose size is different from the neighboring bubbles and showed that the size-differentiated lateral migration in sheared 2D foam can be achieved under some conditions. The key findings can be summarized as the following:

- (a) We introduced a single bubbles whose is different than the neighboring bubbles in a monodisperse foam and found out it migrates laterally as long as the bubble size ratio κ and Ω are both above some thresholds.
- (b) We modified the Chan-Leal theory to account for the observations. The rubbing of large bubbles by the smaller neighboring bubbles resulted into an elevated deformation. After accounting for this fairly high deformation, we were able to predict the migration trajectories of bubbles using the Chan-Leal theory.
- (c) We also explained the presence of thresholds for bubble size ratio and rotational velocity by using a force balance between the wall repulsion and capillary attraction between bubbles.
- (d) And finally lateral migration that was reported in this chapter offers a potential explanation for the size-based segregation in sheared 3D polydisperse foam [28].

Chapter 5

Size segregation in sheared two-dimensional polydisperse foam

5.1 Introduction

As we mentioned in previous chapters, it is widely recognized that complex dynamics of foam is rooted in the foam's microstructure on the bubble scale; the bubbles may undergo breakup, coalescence, coarsening, and morphological changes [2, 6, 17, 21, 68, 76]. Prior experiments have indicated the possibility that bubbles may segregate according to size in a flowing polydisperse foam. But other experiments suggested evidence to the contrary. Herzhaft [28] sheared three-dimensional polydisperse foams between parallel disks, and reported that the large bubbles tend to appear at the middle of the gap while smaller ones are closer to the walls. One explanation is that the bubbles have segregated according to size during the shear. However, an alternative is bubble breakup [27] and coalescence [57] under shear, which could also have produced the observed patterns. In an experiment designed expressly to probe bubble migration, Quilliet et al. [65] produced a monolayer of monodisperse bubbles as a two-dimensional (2D) foam, and inserted a bubble larger than its neighbors. Under oscillatory shear, the large bubble is seen to migrate toward one of the boundaries of the cell. This is inconsistent with Herzaft's report of migration away from walls. In a Hele-Shaw cell, Cantat et al. [5] reported aggregation of large bubbles among smaller neighbors. Cox et al. [15, 24] studied planar extension of bidisperse 2D foams experimentally and numerically, and found no sign of size-based bubble segregation. Therefore, the question of size segregation in flowing polydisperse foam remains open.

For emulsions and suspensions, on the other hand, the segregation and margination of drops and particles in confined flows are well documented [44, 50, 51, 71]. For example, bidisperse suspensions of particles show mild size segregation in 2D channel flow [50, 51]. White blood cells and platelets are found closer to the walls while the red cells aggregate in the center of the tube [44, 71]. We should note, of course, that foams are different from suspensions or emulsions in that the bubbles are closely packed, with relatively little suspending fluid in between. Thus they have a much reduced mobility.

As we showed in previous chapter, we have taken the first step toward answering the question of size segregation in sheared foam by studying the migration of a single large bubble in an otherwise monodisperse bubble raft [55]. In a Couette shear cell, we saw migration of the large bubble away from the walls toward the center of the gap, apparently driven by a "wall repulsion". This appears consistent with the observations of Herzhaft [28] but not those of Quilliet *et al.* [65]. Now in bidisperse and polydisperse foams, a new factor is that the large bubbles interact among themselves as well. How does this interaction affect the migration of the bubbles of different sizes? Do bubbles segregate based on size, and if yes, what is the role of the area fraction of different species? These are the questions we set out to answer in this chapter.

5.2 Size segregation in bidisperse foam

We have done several experiments for bidisperse as well as polydisperse foams. But, it turns out that the key features of size-based segregation are mostly manifested in bidisperse foams already. For the ease of analysis, therefore, we will focus on bidisperse foams in the following, with a final subsection devoted to features specific to polydisperse ones.

Samples		$\Phi_1(\%)$	$\Phi_2(\%)$	$\Phi_3(\%)$	$\Phi_4(\%)$
	А	80	20		
	В	95		5	
	C	90		10	
Bidisperse	D	80		20	
	E	70		30	
	F	50		50	
	G	80			20
	Н	90	5	5	
	Ι	80	10	10	
	J	60	20	20	
	K	40	30	30	
Polydisperse	L	90		5	5
	М	80		10	10
	N	60		20	20
	0	40		30	30

Table 5.1: Composition of the bidisperse and polydisperse foam samples used in the experiments. Φ_1 , Φ_2 , Φ_3 and Φ_4 are the area fractions of the bubble species with radius $a_1 = 350 \ \mu m$, $a_2 = 500 \ \mu m$, $a_3 = 700 \ \mu m$ and $a_4 = 875 \ \mu m$, respectively.



Figure 5.1: Size segregation in sample D under shear ($\Omega = 5$ rpm). The upper row consists of snapshots of the foam at different times: (a) t = 0; (b) t = 2 min; (c) t = 5 min. The lower row shows the corresponding large bubble distributions $\phi_3(y)/\Phi_3$.

Figure 5.1 illustrates a typical process of size-based segregation of the two bubble species under shear. The distribution of the large bubbles, of radius $a_3 = 700 \ \mu m$ in this case, are generated by averaging over several snapshots taken in repeated experiments. In each snapshot, we divide the visible domain of the foam into 9 parallel strips of equal thickness across the gap d, and count the number of large bubbles in each strip. This produces a profile of the area fraction for the large bubbles, $\phi_3(y)$, normalized by the average fraction $\Phi_3 = 20\%$, y being the dimensionless coordinate across the gap with the origin at the center and $y = \pm 0.5$ at the walls. The large bubbles are initially released close to the walls (Fig. 5.1a). Under a rotational rate $\Omega = 5$ rpm, the two species mix at first (Fig. 5.1b). In time, however, the large bubbles aggregate in the center of the gap, within $|y| < y_e \approx 0.25$ in this case, and a quasi-steady state is reach at $t = 5 \min$ (Fig. 5.1c). In this state, there is no statistically significant variation along the azimuthal direction. The quasi-steady distribution of the bubbles is independent of the initial configuration. Figure 5.2 shows that three different initial distributions at the same $\Phi_3 = 20\%$ all lead to the same final distribution. Of course, the time required to reach the final state differs. For brevity, we will refer to the quasi-steady state after prolonged shearing simply as the "steady state".

The apparent aggregation of large bubbles at the center of the gap is consistent with our earlier observations on the migration of single large bubbles in a 2D foam of smaller bubbles [55]. To sum up those findings, a large bubble off the center of the gap experiences an asymmetric "bumping force" from the small bubbles that pass along its sides under shear. This produces a "wall repulsion" toward the center of the gap, much as in the migration



Figure 5.2: Three different initial configurations of sample D (top row), with the large bubbles randomly distributed (a), near the walls (b) and segregated into azimuthal segments (c), lead to the same quasi-steady distribution in the lower row after shearing at $\Omega = 7$ rpm for 10 min. The arrow indicates the direction of shearing.

of a single drop submerged in a suspending liquid [8, 47]. Furthermore, the migration speed can be predicted by the Chan-Leal formula [8] if the capillary number Ca is replaced by an *effective capillary number* Ca_e that is higher than Ca and accounts for the enhanced deformation of the large bubble under the continuous impact of the smaller surrounding bubbles. In the present study, the obvious difference is that there are multiple large bubbles that interact among themselves as well.

There are two prerequisites for the migration of the single large bubble in an otherwise monodisperse foam of smaller bubbles [55]: that the shear rate $\dot{\gamma}$ and the bubble size ratio κ each be above a certain threshold. These reflect the discreteness of the foam; it takes a minimum force to push a large bubble from one row to the next against the capillary pressure in the neighboring bubbles. Such thresholds have also been observed for the bidisperse foams here. In fact, the two threshold values of Ω and κ are expected to be the same as for a single large bubble [55]. Insofar as they are critical values corresponding to the onset of lateral migration of the large bubbles, they are unaffected by the interaction among large bubbles, which arises only after the thresholds have been crossed. For example, no segregation occurs in Sample A for Ω up to 7 rpm, the highest rotational rate possible without incurring centripetal effects [55]; the bubble size ratio $\kappa = a_2/a_1 = 1.43$ is too small. In Sample D ($\kappa = 2$), the threshold is around $\Omega = 3$ rpm. For Sample G ($\kappa = 2.5$), it has come down to around 2 rpm. We have previously presented detailed experimental data on the thresholds [55], along with an analytical expression for the critical condition based on scaling arguments.



Figure 5.3: Steady state distribution of the large bubbles in Sample C after shearing at (a) $\Omega = 3$ rpm and (b) $\Omega = 7$ rpm for 10 min. The solid lines are predictions of the migration-diffusion model to be discussed in subsection III.E.

5.2.1 Effect of shear rate

The shear rate affects both the final steady-state bubble distribution across the gap and the approach to that steady state. Figure 5.3 shows the steadystate distribution of a_3 in Sample C after shearing at different rotational speeds. Evidently, with increasing shear rate the final distribution of the large bubbles becomes more narrowly peaked, and the near-wall regions free of large bubbles widen. For the two cases shown, the half-width of the largebubble distribution $y_e \approx 0.28$ and 0.22 for $\Omega = 3$ and 7 rpm, respectively.

Furthermore, we compare the speed of segregation at different shear rates starting from the same uniform initial configurations. Figure 5.4 plots the temporal evolution of the half-width of the large-bubble distribution, $y_e(t)$. At higher shear rate, the size segregation proceeds at higher speed, and the steady-state distribution is attained within a shorter time. Intuitively, this trend is reasonable. Faster shearing causes more vigorous and frequent impingement of the small bubbles onto the large ones, which should enhance the speed of lateral migration for the latter. A more precise analysis calls for the introduction of another factor, *shear-induced diffusion* of the large bubbles, which influences the steady-state distribution as well.

5.2.2 Effect of area fraction of large bubbles

The size segregation in polydisperse foams differs from the migration of a single large bubble studied before [55] in that the large bubbles interact among themselves. Naturally one expects this interaction to depend on the large-bubble area fraction. By shearing Samples B, D and F, with $\Phi_3 = 5\%$, 20% and 50% for the large bubbles, respectively, we compare the steady-state distributions in Fig. 5.5. By increasing Φ_3 , the distribution becomes



Figure 5.4: Temporal evolution of the half-width of the large-bubble distribution, $y_e(t)$, for Sample D at $\Omega = 3$ and 7 rpm. The solid and dashed lines indicate predictions of the migration-diffusion model.



Figure 5.5: Steady-state distribution of the large bubbles in Sample B, D and F after 10 min of shearing at 7 rpm. These samples have the same bubble sizes but different area fractions for the larger bubbles (see Table 1). The solid lines are predictions of the migration-diffusion model.

broader and the large bubbles are more spread out in the gap. At even higher fractions, the large bubbles become essentially uniformly distributed across the gap.

Moreover, Fig. 5.6 compares the temporal development toward the steady state at two different Φ_3 values. For Sample E at the higher $\Phi_3 = 30\%$, the



Figure 5.6: Temporal evolution of $y_e(t)$ for Samples C ($\Phi_3 = 10\%$) and E ($\Phi_3 = 30\%$) undergoing shear at 7 rpm. The solid and dashed lines indicate predictions of the migration-diffusion model.

equilibrium distribution is achieved more rapidly. In view of the wider distribution in equilibrium (Fig. 5.5c), or equivalently the larger steady-state y_e value, the large bubbles initially near the walls need to travel less distance to reach their equilibrium position. This seems to provide an easy rationalization of Fig. 5.6. But a more careful examination will be made below with the help of a quantitative model.

5.2.3 Effect of bubble size ratio

Figure 5.7 compares the steady-state distributions and temporal evolution of y_e for two bidisperse foam samples with the same large-bubble area fraction Φ but different bubble size ratio (κ). Sample G, with the larger κ , exhibits a more sharply peaked steady distribution, and reaches it more rapidly than Sample D. This mirrors the effects of the shear rate which was explained in 5.2.1. In the migration of a single large bubble in an otherwise monodisperse foam of small bubbles [55], we have found that a larger κ increases the migration velocity as if by elevating the shear rate. In fact, an effective capillary number Ca_e can be defined based on κ that quantitatively captures



Figure 5.7: Effect of the bubble size ratio κ on (a) the steady-state distribution of the large bubbles, and (b) the transient to the steady state in terms of y_e . Sample D has a bubble size ratio $\kappa = 2$ and Sample G has $\kappa = 2.5$. Both have 20% average area fraction for the large bubbles and are subject to $\Omega = 7$ rpm. The solid and dashed lines represent the model predictions.

this effect. The model presented below will make a similar connection for the bidisperse foams.

5.3 A migration-diffusion model

The description above indicates that size segregation in sheared foam is driven by the *migration due to wall repulsion*, the same mechanism as operates on a single large bubble in a medium of smaller ones [55]. A second key player, one that distinguishes the bidisperse foam from the single-large-bubble scenario, is the interaction among the large bubbles themselves. This interaction may be described by the idea of *shear-induced diffusion* that is familiar from prior studies of suspensions and emulsions [22, 34, 41, 48, 50, 51]. The competition between these two factors determines the speed of segregation between bubbles of different sizes and their final distribution.

King and Leighton [41] and Hudson [34] studied the spatial distribution of drops in sheared dilute monodisperse emulsions and investigated the interplay between wall migration and shear-induced diffusion. The evolution of the drop volume fraction ϕ in a simple shear obeys a convection-diffusion equation:

$$\frac{\partial \phi}{\partial t} = -\frac{\partial}{\partial y'} \left(v_m \phi - D \frac{\partial \phi}{\partial y'} \right), \tag{5.1}$$

where v_m is the velocity of wall-induced migration, D is a diffusivity, and y' = yd is the *dimensional* coordinate across the gap. The Chan-Leal for-

mula [8] is used for v_m , in terms of the dimensionless y:

$$v_m = -4\alpha \frac{\sigma}{\eta} \frac{a^2}{d^2} C a^2 \left[y + \frac{8y}{(1 - 4y^2)^2} \right],$$
 (5.2)

where α is a mildly varying function of the drop-to-matrix viscosity ratio given by Chan and Leal [8], σ is the interfacial tension, η is the ambient fluid viscosity, a and d are the drop radius and gap size, and $Ca = \eta \dot{\gamma} a / \sigma$ is the capillary number. The diffusivity D is written as

$$D = \phi \dot{\gamma} a^2 \lambda, \tag{5.3}$$

where λ is a dimensionless coefficient. Balancing the drop fluxes due to wall migration and diffusion, Hudson [34] arrived at the following steady-state profile:

$$\phi(y) = \phi_0 + Pe\left(1 - \frac{y^2}{2} - \frac{1}{1 - 4y^2}\right),\tag{5.4}$$

where $\phi_0 = \phi(0)$ is a constant of integration, and the Peclet number

$$Pe = 4\alpha \frac{a}{d} \frac{Ca}{\Phi\lambda},\tag{5.5}$$

 Φ being the average volume fraction. Note that both v_m and ϕ diverge toward the walls $(y \to \pm 0.5)$. The actual profile comprises the positive central part of Eq. (5.4) and drop-free layers next to the walls, whose edges $(y = \pm y_e)$ are determined by setting $\phi(y_e) = 0$ in Eq. (5.4). Conservation of the drop volume $\int_{-y_e}^{y_e} \phi(y) dy = \Phi$ specifies the centerline volume fraction ϕ_0 .

To adapt this emulsion model to our bidisperse foam, we make the same analogy as was used previously to represent the wall-induced migration of a single large bubble in a sheared monodisperse foam of smaller bubbles [55]. Essentially, we view the smaller bubbles as constituting an effective continuum that suspends and flows around the large bubbles, playing the role of the continuous-phase liquid in the emulsion. Of course, the foam is 2D while the emulsion is 3D, and the smaller bubbles exert a hydrodynamic impact on the larger ones that differs from that of a continuous, viscous liquid. Most importantly, the large bubbles are observed to deform much more than in a viscous liquid under the same capillary number. We previously showed in Chapter 4 that the enhanced deformation can be described by an empirical equation for an *effective capillary number*

$$Ca_e = Ca(2.5\kappa^2 - 7\kappa + 11), (5.6)$$

 κ being the large-to-small bubble size ratio. Ca_e is larger than Ca and, when used in the Taylor formula for drop deformation, predicts the observed bubble deformation. With Caa being replaced by Ca_e , the migration velocity v_m of a single large bubble can be predicted accurately by the Chan-Leal formula [55]. This v_m can be used in the emulsion model (Eq. 5.1) for the bidisperse foam at hand. Then we need only to find the counterpart of the diffusivity of Eq. (5.3).

As far as we know, the idea of shear-induced diffusion has never been used for foams before, and no measured data exist for D or λ . In emulsions, one may consider λ a function of the viscosity ratio, the surface mobility, the capillary number Ca, and the drop fraction Φ . For surfactant-stabilized dilute emulsions, King and Leighton [41] have reported $\lambda(Ca)$ as a weakly rising function of the capillary number Ca (cf. their Fig. 8). In surfactantfree emulsions, Hudson [34] obtained λ values that are an order of magnitude larger, owing to the higher surface mobility. Viewed as an emulsion of the large bubbles in an effective liquid medium, our bidisperse foam is similar to King and Leighton's emulsion in that the surfaces are immobilized by surfactants, and the drop-to-matrix viscosity ratio is negligibly small. Thus, we borrow their dimensionless diffusivity λ , now as a function of the effective capillary number Ca_e . In fact, all our experiments have used low shear rates such that $Ca_e < 0.1$, in which range $\lambda = 0.02 \pm 0.002$ remains essentially constant (see Fig. 8 of King and Leighton [41]). Therefore, we have simply taken $\lambda = 0.02$ in our model calculations. Note that this neglects any dependence of λ on Φ and possibly also on the bubble size ratio κ in our foam. Both prior experiments [34, 41] used dilute emulsions and neither explored the effect of Φ . We assume that λ is independent of the area fraction of the large bubbles for our bidisperse foam. This assumption will be validated a posteriori by comparing the model prediction with experimental data over the whole range of area fraction. With the effective continuum analogy, increasing κ amounts to increasing the effective capillary number Ca_e through Eq. (5.6). As long as we operate in the low- Ca_e regime, the κ effect on λ can be safely neglected.

Having λ thus determined and noting that $\alpha = 81/140$ for an emulsion of negligible drop viscosity [8], we calculate the Peclet number for our bidisperse foam as

$$Pe = \frac{81}{35} \frac{a}{d} \frac{Ca_e}{\Phi\lambda}.$$
(5.7)

With this Peclet number, we can use Eq. (5.4) to predict the steady-state distribution of the large bubbles in our bidisperse foam, and integrate Eq. (5.1) for the transient toward the steady state. In Eq. (5.4), the centerline concentration ϕ_0 is determined from the conservation of drop volume $\int_{-y_e}^{y_e} \phi \, dy = \Phi$. Equation (5.1) is integrated using finite difference with boundary conditions $\phi(\pm y_e) = 0$, y_e being determined iteratively from the drop volume conservation by the shooting method. Both the steady ϕ profile and the transient can be compared with measurements. In particular, we will examine the effects of the shear rate $\dot{\gamma}$, the average area fraction Φ and the bubble size ratio κ .

Figure 5.3 compares the model predictions with the measured steadystate distributions for the bidisperse Sample C at two shear rates, and Fig. 5.4 compares the temporal development of the distribution for Sample D. In both cases, the rotational speed of 3 and 7 rpm correspond to capillary numbers $Ca = 5.8 \times 10^{-3}$ and 1.4×10^{-2} , which in turn correspond respectively to effective capillary numbers $Ca_e = 4.0 \times 10^{-2}$ and 9.5×10^{-2} . Based on these parameters, the predicted steady-state profile and its temporal development are both in reasonably good agreement with experimental measurements. With increasing shear rate, the large bubbles migrate away from the walls more rapidly, and this aggregation at the center overpowers the shear-induced diffusion that strives to spread the large bubbles uniformly. Consequently, the size-based segregation occurs more rapidly for higher shear rates, and produces a narrower equilibrium distribution centered at the middle of the gap y = 0. Note that Eq. (5.1) does not predict a $t \sim \dot{\gamma}^{-1}$ scaling for the transient. It would if v_m and D were both proportional to $\dot{\gamma}$ or Ca. In reality, $v_m \propto Ca^2$, and D also depends on Ca nonlinearly thanks to $\lambda(Ca)$ [41]. Our experimental data do not exhibit such a scaling either.

As the average area fraction of the large bubbles Φ_3 increases, Fig. 5.5 shows that the model correctly predicts the widening of the equilibrium distribution, and the agreement with measurements is quantitatively accurate. The idea underlying this prediction is that higher fraction of the large bubbles increases the frequency of their collision and thereby elevates the effective diffusivity D (cf. Eq. 5.3). This has been confirmed by the experiments. We have also studied the effect of area fraction on the speed of size segregation. The model predicts that with increasing Φ_3 , the segregation occurs more rapidly (Fig. 5.6); it takes less time to reach the equilibrium distribution. This captures the trend in the experimental data if not the precise values of the segregation time. Qualitatively, increasing Φ_3 increases the diffusivity D, which should counteract the migration and lead to a slower segregation. On the other hand, a higher Φ_3 corresponds to a wider equilibrium distribution with a larger y_e . This means that large bubbles initially near the wall need to travel a shorter distance to get to their steady-state position. These two effects oppose each other and the outcome seems to be in favor of the latter. King and Leighton [41] have quantified the competition between the two effects in the limiting case of small y. By linearizing the migration velocity v_m of Eq. (5.2) (i.e., reducing the y terms between the brackets to 9y), they obtained a self-similar solution in which time t scales only with d/v_m , and is independent of Φ_3 . Our experiment and analysis are not restricted to the small-y limit, and thus do not exhibit the similarity. Recall that we have assumed λ to be independent of Φ in Eq. (5.7). The close agreement for the whole range of area fractions studied here indicates that this is a reasonable assumption.

Finally, we examine the effect of the bubble size ratio κ , which influ-



Figure 5.8: Steady-state bubble distributions in the polydisperse foam samples H, I, J and K, after 10 min of shearing at 7 rpm. The samples have the same three bubble sizes, $a_1 = 350 \ \mu m$, $a_2 = 500 \ \mu m$ and $a_3 = 700 \ \mu m$, at different average area fractions: (a) Sample H, $(\Phi_1, \Phi_2, \Phi_3) = (90\%, 5\%, 5\%)$; (b) Sample I, (80%, 10%, 10%); (c) Sample J, (60%, 20%, 20%); (d) Sample K, (40%, 30%, 30%). The area fractions are normalized for each species.

ences the structural evolution of bidisperse foams through the $Ca_e \sim Ca$ relationship in our model (Eq. 5.6). Figure 5.7 shows that the model correctly predicts the effects of κ on the steady-state distribution as well as on the temporal evolution toward it: higher κ produces a faster approach to a narrower steady-state distribution. Qualitatively and quantitatively (via Eq. 5.6), therefore, increasing κ has similar effects to elevating the shear rate or capillary number.

5.4 Polydisperse foam

We now consider polydisperse foams composed of three bubble sizes, a_1 , a_2 and a_3 for Samples H–K and a_1 , a_3 and a_4 for Samples L–O (see Table 1). Note that in these samples the two larger species always have the same area fraction. Figure 5.8 shows the steady-state distributions for Samples

H–K. For Sample H with the lowest Φ_3 , the largest bubbles (of radius a_3) exhibit a sharply peaked distribution at the center of the gap while the two smaller bubble species (a_1 and a_2) are more or less uniformly distributed. If the a_3 bubbles were absent, the a_1 and a_2 bubbles would not exhibit size segregation as their size ratio $\kappa = 1.43$ is below the threshold for $\Omega = 7$ rpm [55]. Therefore, the aggregation of the a_3 bubbles in Sample H is similar to that in a bidisperse foam. By increasing the area fraction Φ_2 and Φ_3 to 10% and 20% (Samples I and J), the two smaller bubble species are displaced toward the walls. This is evidently due to the increasing area occupied by the a_3 bubbles at the center, and recalls the marginalization of white blood cells when the more flexible red cells aggregate in the center [44, 71].

However, increasing Φ_2 and Φ_3 further to 30% (Sample K) brings about an apparent *reversal* of the marginalization. Now all three species are roughly uniformly distributed in the gap. This can be rationalized by the stronger shear-induced diffusion of the largest bubbles at the higher Φ_3 , much as in the bidisperse foams of Fig. 5.5. Comparing Fig. 5.8 and Fig. 5.5, however, reveals an interesting role for the a_2 bubbles. In Fig. 5.8(d), the a_3 distribution flattens for $\Phi_3 = 30\%$ in the polydisperse Sample K, whereas in the bidisperse Sample F (Fig. 5.5c), the large bubbles are not quite uniformly distributed even for $\Phi_3 = 50\%$. Thus, the a_2 bubbles are not inert and merely passively displaced by the a_3 bubbles. They actively facilitate the spreading of the largest bubbles. This may have occurred through hindering their migration toward the center (via effectively reducing κ) or enhancing the diffusion of the largest bubbles, or even both.

Now we investigate the size segregation in polydisperse Samples L–O in which the two larger species, a_3 and a_4 , both tend to migrate away from the walls and compete with each other to occupy the center of the gap. Figure 5.9 shows the equilibrium distributions of the three bubble species subject to shearing at $\Omega = 7$ rpm. As it turns out, the two large bubble species behave similarly in this case. For $\Phi_3 = \Phi_4 \leq 20\%$ (Samples L– N), both a_3 and a_4 bubbles aggregate at the center of the gap. The a_1 bubbles are marginalized as seen above. The largest a_4 species enjoys a narrower distribution with a higher peak than a_3 . Thus, the larger bubble size κ affords the former an advantage. With increasing Φ_3 and Φ_4 , the distributions broaden until at 30%, both become more or less uniformly distributed across the gap (Sample O). As in Fig. 5.5(c) and Fig. 5.8(d), this can be ascribed to the dominance of the shear-induced diffusion of the a_3 and a_4 bubbles.

5.5 Conclusion

We have studied the structural evolution of bidisperse and polydisperse 2D foams in a narrow-gap Couette shear cell. Within the parameter ranges



Figure 5.9: Steady-state bubble distributions in the polydisperse foam Samples L, M, N and O, after 10 min of shearing at 7 rpm. The samples have the same three bubble sizes, $a_1 = 350 \ \mu\text{m}$, $a_3 = 700 \ \mu\text{m}$ and $a_4 = 875 \ \mu\text{m}$, at different area fractions: (a) Sample L, $(\Phi_1, \Phi_3, \Phi_4) = (90\%, 5\%, 5\%)$; (b) Sample M, (80%, 10%, 10%); (c) Sample N, (60%, 20%, 20%); (d) Sample O, (40%, 30%, 30%). The area fractions are normalized for each species.

tested, the main experimental findings can be summarized as follows.

- (a) After shearing for a sufficiently long time, the foam achieves a quasisteady morphology that is independent of the initial configuration.
- (b) In this quasi-steady state, the bubble species may be uniformly mixed or segregated by size depending on the physical and flow parameters. Size segregation occurs if the bubble size ratio and shear rate are both above certain threshold values, and if the area fraction of the large bubbles is not too high. Otherwise a mixed state obtains.
- (c) In size-segregating bidisperse foams, the segregation occurs more rapidly and produces a narrower final distribution for higher shear rates and larger bubble size ratios. Increasing the area fraction of the large bubbles, on the other hand, leads to a broader final distribution that is achieved in less time.

(d) Polydisperse foams behave similarly in that size segregation occurs at relatively low area fractions of the largest bubbles while a uniformly mixed morphology prevails at higher large-bubble area fractions. The bubbles of intermediate size tend to facilitate the broadening of the distribution of the largest bubbles.

These observations are rationalized by adapting a migration-diffusion model previously developed for monodisperse emulsions. Viewing the larger bubbles as being suspended in an effective continuum comprising the smaller ones, we describe the structural evolution in bidisperse foams by a convectiondiffusion equation. The model balances two competing factors, the lateral migration due to wall repulsion and the shear-induced diffusion due to interaction among the large bubbles. For bidisperse 2D foams, the model predicts all aspects of the experimental observations, often with quantitative accuracy.

The success of the emulsion model in predicting bubble segregation in a polydisperse bubble raft is quite remarkable, especially in view of the differences between the two systems. The prevailing thinking of foam dynamics is that it is determined by the interfacial morphology on the local scale. Then the 2D foam studied here can be viewed as a curious exception where at least one attribute of the dynamics, the migration and segregation of bubbles based on size, turns out not to be intimately related to the morphology of the smaller bubbles. These small bubbles can be replaced, in a sense, by an effective continuum while preserving the same segregation of the large bubbles. There are some caveats to this analogy, however. The "replacement" of the surrounding bubbles by an effective continuum is so as to produce the same amount of deformation on the large bubbles. This boils down to an effective capillary number. One cannot reduce the analogy further to something more tangible, say an effective viscosity, which would not produce the correct migration velocity from the Chan-Leal formula (Eq. 5.2). Thus, the effective capillary number embodies intricate local dynamics having to do with the discreteness of the surrounding bubbles, which exert a "bumping force" on the large bubbles [55] that cannot be ascribed to an elevated medium viscosity. Moreover, the analogy may be limited to certain types of foam. In our experiment, the bubbles are closely packed but not pressed against one another so as to produce polygonal facets. If we try to pack more bubbles into the raft, they tend to pile on top of others and destroy the two-dimensionality. Thus, the smaller bubbles are essentially undeformed in our experiments. In drier foams that undergo more intensive interaction among bubbles, e.g. through T1 events [79], the continuum analogy may no longer hold.

To conclude, let us briefly return to prior experiments that motivated our study. Our findings suggest that in the prior experiment of Herzhaft [28], where 3D polydisperse foams are sheared between parallel plates, shear-

5.5. Conclusion

induced migration probably have occurred to produce marginalization of smaller bubbles to the plates and a central layer rich in large bubbles. However, three-dimensionality affects how neighboring bubbles interact with one another, and our 2D model will need to be upgraded before it can be compared quantitatively to 3D foam experiments. In addition, it is important to note the experimental and numerical results of Cox and coworkers [15, 24] that showed no size segregation in 2D foams undergoing cyclic planar extension and compression. The conditions in their studies differ from ours in at least three aspects. In extensional flows the bubbles do not follow parallel streamlines. Instead, neighboring rows are compressed into one while being elongated in the orthogonal direction. Thus, the interaction among bubbles differs markedly from the rubbing and bumping in our shear experiments. Moreover, the cyclic straining introduces repeated encounters among bubbles, a feature absent from steady shearing. Finally, the maximum extensional rate in their experiments is only 0.0455 s^{-1} , much below our threshold value of 4.17 s^{-1} for approximately the same bubble-size ratio $\kappa = 2$. Their simulation employed the Surface Evolver, and is thus quasistatic in nature. It appears, therefore, that size segregation in extensional flows remains an open question that requires further studies, especially at high strain rates.

Chapter 6

Effect of non-Newtonian rheology on bubble migration in sheared foam

6.1 Introduction

Foams are quintessential soft matter in that they admit both a macroscopic, continuum-based description and a microscopic, bubble-scale one. On the one hand, foam rheology is invariably measured on the bulk. In so doing, one implicitly adopts an effective continuum view, and sometimes explicitly represent the foam rheology by continuum models [39, 46]. On the other hand, bulk flow and deformation produces changes of the microstructure, i.e. bubble-scale morphology. Shearing is known to induce neighbor-swapping rearrangements known as T1 processes [62, 77]. Additional microstructural changes include bubble coalescence, breakup, migration and size-based segregation [27, 28, 56, 57]. Since foams can be examined on both levels, and indeed manifest a clear link between their microstructure and bulk flow behavior, they are excellent model systems for studying the coupling between the microscopic and macroscopic scales.

We are just beginning to understand the interaction between the two length scales, and many questions remain to be answered. Even the shear viscosity of a foam is not well understood. In one simple-shear experiment, Golemanov *et al.* [27] observed a marked increase of the shear stress in time after the start of shear, and attributed it to the breakup of the bubbles. In another experiment, Herzhaft [28] reported a shear stress that gradually declines in time. In addition, foams show shear-thinning, which can be fitted to the continuum Herschel-Bulkley model [39, 46]. Surprisingly, the bubble velocity profile under shear differs appreciably from that predicted by the continuum model [39]. This has been ascribed to a nonlocal effect arising from the cooperative movement of bubbles within a certain "cooperativity" length scale. Surface tension is also known to produce normal stress differences on the macroscopic scale [25, 45]. Thus, the micro-macro connection is subtle for foams, and their dynamics is influenced by continuum rheology as well as textural granularity.

We have been investigating another aspect of this connection, through

the cross-streamline migration of bubbles in sheared "two-dimensional" foam, which is a bubble raft floating on a soapy solution. The two-dimensionality affords direct visualization of bubble-scale microstructures that would be impossible for 3D foams. As we showed in the previous chapter, bubbles segregate according to size in sheared polydisperse foams, and that the segregation can be understood based on a simple continuum model in which the smallest bubbles are viewed as an effective Newtonian fluid that suspends the larger bubbles. In this model, combining shear-induced migration of individual bubbles and an effective diffusion due to collision among large bubbles can account for the segregation data very well. This adds to the collection of foam behavior that can be described as continuum. In the mean time, the discreteness of the bubbles manifests itself as well, in terms of "quantized" steps of migration and thresholds in shear-rate and bubble size ratio under which no migration takes place. Note that the above has been observed in simple shear in a narrow-gap Couette device.

The experiments to be presented in this chapter extends the above study to nonuniform shear in a wide gap Couette device. This seemingly naive change of geometry, as it turned out, brings out the non-Newtonian rheology of the foam to bear on the migration of bubbles. Therefore, this may be viewed as an interesting example of the bulk rheology affecting the microstructural evolution of the foam. Bubble migration in foam can be contrasted with drop migration in non-Newtonian fluids, a subject that has received long-standing attention [8, 9, 26]. As will be demonstrated, bubble migration in a non-Newtonian liquid holds the key to understanding the migration in foam. In fact, the continuum analogy can be maintained if we view the smaller bubbles as constituting an effective non-Newtonian fluid that shows shear-thinning and normal stress difference under shear.

In quasi-static state liquid foam shows topological changes known as T1 and T2 events[21, 46, 62]. Furthermore, bubbles in liquid foam may undergo breakup, coalesce or even segregate according to the size when subject to high shear rates [27, 28, 56, 57]. In chapter 4 and chapter 5 in narrow gap, we explored the structural change due to size-based bubble segregation, and found that the surrounding bubbles can be viewed as an effective Newtonian continuum in some sense[55, 56].

In this chapter, we show how the continuum idea can be extended to a situation where the foam must be seen as a non-Newtonian fluid. Therefore, the main objective of this work is to study the non-Newtonian behavior of liquid foam and its effect on structural evolution. As will be demonstrated, bubble migration in a non-Newtonian liquid holds the key to understanding its migration in foam. Studies on migration of a single droplet in non-Newtonian fluids show that the migration differs from the Newtonian fluid [8, 9, 26]. The main objective in this chapter is to understand the migration in foam through the window of non-Newtonian effects. Therefore, we make some well characterized viscous and elastic fluids that represent the

shear-thinning and normal stresses in sheared foam and study the migration process in them.

6.2 Experimental results

We have performed two series of experiments—bubble migration and rheological measurements—on three types of mediums: two-dimensional foams, xanthan gum solutions and a Boger fluid. In the following, we present the result of each experiment in turn.

6.2.1 Bubble migration in foam

The experimental protocol for recording bubble migration across streamlines is similar to that used in narrow-gap Couette cells in chapter 3. We make a monodisperse foam consisting of bubbles of radius $r = 0.36 \pm 0.02$ mm, which covers the entire wide gap of the Couette device in a more or less regular hexagonal lattice. The foam quality, defined as the area fraction of the bubbles, is maintained at 85% for all the experiments to be presented. We then insert a single large bubble of radius R into the foam at different initial positions, and shear the foam by rotating the inner cylinder at a constant angular velocity Ω . The two control parameters are the nominal shear rate and the bubble size ratio $\kappa = R/r$. Shear rate at the inner wall can be estimated from from the velocity gradient that the first row of bubble experiences at each rotational speed. Therefore, shear rate $\dot{\gamma}$ varies from 1.5 to 8.62 s⁻¹; the upper bound is chosen such that centripetal force remains negligible in all experiments. Foams in our wide-gap device may yield partially, and that introduces unnecessary complication to the discussion of migration. Thus, from here onward, we will only consider shear rates above that required for full yielding: $3.5 \text{ s}^{-1} < \dot{\gamma} < 8.62 \text{ s}^{-1}$. For the large bubble we have tested five sizes: R = 0.5, 0.6, 1, 1.4 and 1.8 mm, corresponding to $\kappa = 1.39, 1.67, 2.79, 3.91$ and 5.03.

Similar to what has been reported for the narrow-gap Couette device in chapter 4, the large bubble migrates across the flow direction if the shear rate $\dot{\gamma}$ and bubble size ratio κ are each above a threshold value. The migration is driven by a hydrodynamic force that arises from the asymmetric flow and pressure fields surrounding the deformed bubbles [55, 56]. The thresholds reflect the discreteness of the foam; the hydrodynamic force has to overcome the capillary pressure in neighboring bubbles in order to move the large bubble to the next row. The migration is generally away from the walls, and the hydrodynamic driving force is greatest at the wall and diminishes toward the center [8]. Thus, a large bubble may migrate across one or several rows if released near the wall, but not at all if released further away from the wall. For simplicity, we will exclude such partial migration from further discussion, and define the thresholds of $\dot{\gamma}$ and κ according to complete migration, i.e.

migration to an equilibrium position regardless of initial positions. As in the narrow-gap Couette cell, we find the $\dot{\gamma}$ threshold to decrease with increasing κ , and the κ threshold to decrease with increasing $\dot{\gamma}$ [55]. The threshold values are comparable to those in the narrow gap. In the following we will concern ourselves only with the dynamics above these thresholds.



Figure 6.1: Migration trajectories of bubbles of two size $R = 1 \text{ mm} (\kappa = 2.79)$ and $R = 1.4 \text{ mm} (\kappa = 3.91)$, released from different positions in the foam sheared at different shear rates. The bubble center is given by s, its distance from the inner cylinder scaled by the gap width $d = R_o - R_i$. The curve shows Chan-Leal's prediction for the bubble size R = 1.4 mm undergoing shear rate of $\dot{\gamma} = 5.71 \text{ s}^{-1}$.

Figure 6.1 shows the migration trajectories of bubbles of two sizes ($\kappa = 2.79$ and $\kappa = 3.91$) at different shear rates. The threshold shear rate is around 5.71 for the smaller bubble, and around 3.5 for the larger one. For shear rates above this threshold, the final equilibrium position is reached from all initial positions. This equilibrium position seems to be independent of the shear rate and the bubble size ratio, although the speed of migration

increases with $\dot{\gamma}$ and κ . The features described so far are similar to prior observations in the narrow-gap Couette device [55].

The key difference is that the equilibrium position for large $\dot{\gamma}$ is not at the center of the gap, as is the case in the narrow-gap geometry [55]. Rather it is some distance inward from the center of the gap, closer to the inner cylinder; in the particular case shown in Fig. 6.1, this position is at $s \approx 0.36$. One naturally seeks a geometrical explanation for the difference. After all, the wide-gap Couette device should produce a nonuniform shear rate profile across the gap, with higher local shear rate in the inner half the gap than the outer half. This asymmetry should bias the equilibrium position of the migrating bubble. This effect can be quantified with the help of the Chan-Leal theory for lateral migration of droplets in Couette flows [8, 9].

Although the Chan-Leal formula was developed for the migration of a single drop in a continuum suspending fluid, we have demonstrated that it can accurately describe the migration of a single large bubble in a sea of monodisperse bubbles if the enhanced bubble deformation is accounted for through an elevated effective capillary number [55, 56]. Since the Chan-Leal theory was developed for a Couette device, it accounts for the curvature in the streamlines and the variation of shear rate across the gap. Thus we have used the *modified* Chan-Leal formula, containing the effective capillary number, to predict the migration of a single bubble corresponding to the conditions of one of the experimental runs of Fig. 6.1, and the result is plotted as a solid curve. It predicts only a slight inward shift of the final equilibrium position, s = 0.47, which cannot account for the much larger shift observed experimentally. Besides, the migration speed is also overpredicted by a wide margin. Therefore, the observations in the wide-gap experiment cannot be accounted for by the geometry alone.

A factor that has not been taken into account in the above comparison is the non-Newtonian features of the liquid foam. The Chan-Leal formula used in Fig. 6.1 is for a Newtonian suspending fluid. It has successfully represented the migration observed in our previous experiments [55, 56], which implies that the small-bubble foam can be viewed effectively as a Newtonian suspending fluid. Can it be that the nonuniform shearing in the wide-gap device brings out non-Newtonian rheology that is not manifest in the narrow-gap Couette cell? Shear-thinning will accentuate the nonlinearity of the velocity profile, and a large bubble would thus experience unequal viscosities upon its two sides. Moreover, the first normal stress difference N_1 would also exhibit an asymmetry between the two sides. To ascertain these potential effects on bubble migration in the foam, we need to characterize the bulk rheology of the foam first. As will become clear in the next subsection, this has in turn motivated us to make polymer solutions possessing shear thinning and elasticity separately, in which bubble migration may be investigated as benchmarks for gauging the bubble migration in foam.



Figure 6.2: Shear flow curve of the two-dimensional foam measured in a rheometer with a bob-cup fixture. Two data sets are plotted along with a best-fitting curve to the Herschel-Bulkley equation (Eq. 6.4).

6.2.2 Bulk rheology of 2D foam and polymer solutions

Foams are known to have a yield stress, and in the fully yielded state exhibit shear-thinning and normal stress differences [25, 39, 42, 60]. To probe the shear-thinning of our 2D foam, we have measured its shear rheology on a rotational rheometer using the bob-cup fixture. To accommodate a large number of bubbles, we used a wide-gap setup, with the radius of the inner cylinder being 22 mm and that of the stationary outer cylinder being 35 mm. The local shear rate at the inner cylinder is obtained from the following [23, 43]:

$$\dot{\gamma} = 2 \Omega \frac{d(\ln\Omega)}{d(\ln M)}.$$

$$\mathbf{If} \left(\sigma_o \leq \sigma_y \leq \sigma_i\right) \quad (6.1)$$

$$\dot{\gamma} = 2 \frac{d(\ln\Omega)/d(\ln M)}{1 - R_i^2/R_o^2} - \frac{\Omega - M(d\Omega/dM)}{\ln(R_i/R_o)}.$$

$$\mathbf{If} \left(\sigma_o > \sigma_y\right) \quad (6.2)$$



Figure 6.3: Shear viscosity of xanthan gum solutions of various concentrations. The line indicates the foam viscosity.

 $\sigma_o, \sigma_i, \sigma_y$ and M being the shear stress at the outer cylinder, shear stress at the inner cylinder, the yield stress and torque at the inner cylinder respectively. The shear rate can be estimated from the maximum of equations(6.1, 6.2). According to Estelle *et al.* [23] the appropriate shear rate is the one that maximizes the dissipation in flowing material.

Figure 6.2 shows the shear stress as a function of the shear rate for our 2D foam. Following prior experiments on 2D and 3D foams [39, 60], we fit the data by a Herschel-Bulkley model:

$$\sigma = \sigma_y + K \dot{\gamma}^n, \tag{6.3}$$

with a yield stress $\sigma_y = 0.32$ Pa, consistency K = 0.77 Pa·sⁿ and a powerlaw index n = 0.47. Thus, our foam shows similar shear-thinning behavior to previous experiments [39, 60]. Both 2D and 3D foams are known to exhibit a first normal stress difference N_1 [25, 42, 45, 59]. Labiausse *et al.* [45] measured N_1 for a 3D foam in the pre-yielding regime. Kraynik *et al.* [42] further determined that N_1 is on the same order of magnitude as the shear stress for 3D foam before yielding. In simulations of a random 2D foam


Figure 6.4: Comparison of the power-law viscosity of xanthan solutions of different concentrations (symbols) with that of the foam (horizontal lines). (a) The power-law index n; (b) the consistency factor K.

undergoing simple shear in the yielded regime, Okuzono *et al.* [59] recorded N_1 values roughly twice as large as the shear stress over a range of shear rates. For our 2D foam, we have not been able to measure N_1 directly. In view of the limited data in the literature, we have decided to use the results of Okuzono *et al.* [59] as a guideline, and assume that for our 2D foam N_1 is on the same order of magnitude as the shear stress, which we have measured with confidence.

As shear-thinning and normal stress act simultaneously on bubble migration in our foam, it is impossible to identify and analyze their individual contributions. Therefore, we have sought to probe the two effects separately by using shear-thinning and viscoelastic polymer solutions that represent each aspect of the foam's rheology.



Figure 6.5: Shear rheology of the Boger fluid, with open circles for the shear viscosity and filled diamonds for the first normal stress difference N_1 . The straight line is a power-law fitting for N_1 with a slope close to 2. The filled squares show the shear stress of the foam, which is comparable in magnitude to N_1 of the Boger fluid, especially near the upper bound of the shear rate.

Aqueous solution of xanthan gum are known to exhibit shear thinning but negligible elasticity [1, 3]. We have tested a series of xanthan solutions and chosen the closest one to the foam rheology. Figure 6.3 shows the shear viscosity of xanthan solutions of 6 concentrations. For comparison, we have plotted the viscosity of our foam in the range of shear rates encountered in the bubble-migration experiments. Furthermore, we fit a power-law to the xanthan viscosities in the same range, and plot in Fig. 6.4 the consistency K and the power-law index n for the xanthan solutions together with the values measured for our foam. The closest one to the foam appears to be the solution at a concentration of 2500 ppm. Therefore, we choose this solution as the representative for the shear-thinning behavior of foam.

Similarly, Fig. 6.5 compares the shear rheology of the Boger fluid with that of the foam. Within the range of shear rates tested, the Boger fluid exhibits an essentially constant shear viscosity, and an N_1 that scales approximately with $\dot{\gamma}^2$. Ideally, we would have liked N_1 of the Boger fluid to match the foam shear stress in the $\dot{\gamma}$ range of interest, up to 6 s⁻¹. This turns out to be difficult to realize experimentally. For one, increasing the polymer concentration in the Boger fluid brings forth appreciable shear thinning. Thus, we have accepted this Boger fluid as roughly representing the order of magnitude of the normal-stress in the foam.

6.2.3 Bubble migration in shear-thinning and Boger fluids

We have conducted bubble migration experiments in the shear-thinning xanthan solution and the Boger fluid, using the same wide-gap Couette device, bubble sizes and operating conditions as in the foam experiments. Figure 6.6 depicts migration of large bubbles of two sizes in the xanthan solution at different shear rates and initial positions. Our results show that the same equilibrium position, $s \approx 0.25$, is reached from different initial positions. This position is roughly midway between the center of the gap and the inner cylinder, and is much more inward than that in a Newtonian fluid (s = 0.47 according to the modified Chan-Leal formula; see Fig. 6.1). Besides, the equilibrium position does not depend on the bubble size, nor on the shear rate. But the speed of migration does increase with the bubble size and the shear rate. Thus, shear-thinning tends to shift the bubble further toward the inner cylinder. This conclusion is consistent with the previous experimental results of Gauthier et al. [26]. In a wide gap Couette device, Gauthier et al. studied migration of a deformable droplet in a shear-thinning fluid with power-law index n = 0.71. Droplets of different sizes starting from different initial positions all end up at an equilibrium position $s \approx 0.4$.

Our xanthan solution has stronger shear-thinning (n = 0.43) than their fluid, and it is reasonable that the bubbles assume a position farther inward than in their case. Regarding the hydrodynamic origin of the effect, one may imagine that the bubble experiences reduced viscosity on the side closer to the inner wall, where the shear rate is higher. This may have biased the lateral force in favor of inward migration. However, such a naive argument fails to anticipate the apparent insensitivity of the equilibrium position to $\dot{\gamma}$ and κ . In the next subsection, we will use numerical simulations of drop migration in shear-thinning fluids to explore these questions.

The opposite trend is observed in the Boger fluid. Figure 6.7 shows typical migration trajectories of bubbles of three sizes, released at different initial positions, at two shear rates. In all cases, the bubble migrates to an equilibrium position close to s = 0.55 in the outer half of the gap. This



Figure 6.6: Migration trajectories of bubbles in the xanthan solution starting from different initial positions. The bubbles are of two sizes R = 0.6 mm and R = 1 mm, and the shear rate is varied from $\dot{\gamma} = 3.5 \text{ s}^{-1}$ to $\dot{\gamma} = 8.62 \text{ s}^{-1}$.

suggests that the normal stress N_1 tends to force the bubble outward. Furthermore, the equilibrium position shows no dependence on the shear rate $\dot{\gamma}$ and little dependence on the bubble size κ , although the migration speed increases with both.

Chan and Leal [8, 9] have carried out experimental and theoretical studies of the migration of a single suspended drop in a Boger fluid sheared in a Couette device. The predictions of the Chan-Leal formula, for the experimental conditions used here, are plotted as solid and dashed lines in Fig. 6.7. First, the formula correctly predicts the outward shift of the bubble's equilibrium position in all cases. Second, for a fixed bubble size, the formula predicts a final equilibrium position that is independent of the shear rate, in agreement with our observations. In Fig. 6.7(a), the predicted $s \approx 0.6$ differs from our measurement by 8%. This difference in the destination affects the prediction of the migration speed, but it still falls within reasonable agreement with experimental data. Third, the Chan-Leal formula predicts the equilibrium position s to shift away from the outer wall as the bubble



Figure 6.7: Migration trajectories of a single bubble in the Boger fluid. (a) A bubble of radius R = 1 mm released from two initial positions at two shear rates. (b) The effect of bubble size at a fixed shear rate ($\dot{\gamma} = 4.77 \text{ s}^{-1}$). The curves show the predictions of the Chan-Leal formula.

size R increases relative to the gap $d = R_o - R_i$. This effect is stronger for smaller R/d and saturates for larger R/d. In comparison, our experimental data shows a much weaker effect of bubble size. As R increases from 0.7 mm to 1 mm and then to 1.4 mm, s seems to shift slightly inward toward the centerline, but the magnitude is much below the roughly 10% change in s predicted by the Chan-Leal formula.

6.3 Discussion

In this section we will try to understand the underlying physics behind the experimental results. To do so, we will strive to answer three important questions separately: (a)- Why does elasticity (N_1) shift the equilibrium position outward? (b)- Why does shear thinning have an opposite effect? (c)- What is the effect of combination of these two factors?

6.3.1 Effect of elasticity

We start with the first question: how does elasticity (N_1) shift the equilibrium position outward? Karnis and Mason [37] suggested that a particle in a Boger fluid experiences normal forces due to N_1 on both of its sides. In the nonuniform shear of a wide-gap Couette device, the normal force is larger on the inner side as the shear rate is higher there. This asymmetry pushes the droplet toward regions of lower shear rates, i.e. toward the outer wall. Ho and Leal [29] introduced the idea of "hoop thrust" to rationalize the N_1 effect on particle migration. They suggested that the presence of the particle disturbs the flow field around it and generates "bowed streamlines" around the particle. The tension along these streamlines thus produce a hoop stress that tends to drive the particle toward regions of lower shear rates. Both explanations are similar in essence; the gradient in shear rate leads to a gradient in normal stress, which then pushes the particle toward the outer wall.

Another intriguing feature of Fig. 6.7(a) is the independence of the equilibrium bubble position to the shear rate $\dot{\gamma}$. There are two mechanisms governing migration of a deformable particle in a Boger fluid, the normal stress as discussed above, and the deformation of the bubble or drop. As demonstrated by Chan and Leal [8] in Newtonian as well as second-order fluids, the bubble deformation creates an asymmetry in the flow and stress fields in the vicinities, which tends to push the bubble away from solid walls. Thus, this effect is opposite to that of N_1 in the outer half of the Couette device. When the shear rate $\dot{\gamma}$ is elevated, N_1 increases and so does the bubble deformation. The Chan-Leal theory shows that these two effects cancel out such that the final equilibrium position is independent of $\dot{\gamma}$. This has been born out by our experimental data as well (Fig. 6.7a). Our experimental conditions also satisfy the constraints under which the Chan-Leal asymptotic theory holds. These conditions are $Ca \ll 1$, $R/d \ll 1$, $De \ll 1$ and De << Ca.

The effect of the bubble size R can be considered in a similar way. A larger bubble will experience larger deformation on the one hand, and a larger N_1 -based normal force on the other. Again the two factors tend to oppose each other. The Chan-Leal calculation shows, however, that they do not exactly cancel each other. Smaller bubbles tend to favor the N_1





Figure 6.8: Calculated force for Boger fluid and Newtonian fluid vs radius on a bubble with radius of 1 mm in wide-gap Couette geometry using Chan-Leal formula.



Figure 6.9: Dimensionless equilibrium position vs dimensionless radius of the bubble in Boger fluid sheared at $\dot{\gamma} = 4.77 \ sec^{-1}$. Curve shows the prediction of Chan-Leal's model and the data are experimental results.

effect, and thus attain an equilibrium position closer to the outer wall. With increasing bubble size, the equilibrium position shifts inward but levels off for $R/d \approx 0.11$ (c.f. Fig. 6.9). The experimental data, covering a modest range of R/d, do show a clear downward trend, although the slope is not as steep, and the equilibrium positions are generally farther from the outer cylinder than the theoretical prediction. The largest discrepancy is for the smallest drop (R = 0.7 mm). At present we have no explanation.

We can rationalize the migration results of a bubble in Boger fluid by estimating the lateral force on the bubble during the migration process. The wall repulsive force can be computed from the Stokes formula using the migration velocity of the Chan-Leal theory for both Newtonian and Boger fluids. Fig 6.9 shows calculated force vs dimensionless position in different fluids. For a Newtonian fluid this force goes to zero at $s \approx 0.48$ which indicates the equilibrium position of a migrating bubble and for similar bubble in Boger fluid equilibrium position shifts toward the outer cylinder $s \approx 0.6$. This confirms our experimental results on migrating bubbles in Boger fluid.

6.3.2 Effect of shear thinning

Shear thinning behavior of foam is an important non-Newtonian feature that might have affected the migration of bubble under shear. To the best of our knowledge, there is no theory that can explain the effect of shear thinning on lateral migration of a deformable object. Therefore, we have carried out several 2D simulations using finite element method. Simulations would allow us to analyze the bubble trajectory and force on a bubble inside a shear thinning fluid. The shear thinning parameters were chosen to represent the xanthan gum solution with following parameters: n = 0.43, $K = 1 \ (Pa.s^{0.43}), 0.025 \le R/d \le 0.1 \text{ and } 1.37s^{-1} \le \dot{\gamma} \le 11s^{-1}$ in a Couette co-axial cylinders geometry with $R_i = 2 \ cm$ and $R_o = 3 \ cm$. The size of computational domain is different from the one in experiments ($R_i = 8.1 \ cm$ and $R_i = 9.9 \ cm$). This is mainly due to the technical difficulties in simulating of very large domains.

Fig.6.10 shows the trajectory of a migrating bubble in shear thinning fluid obtained by simulations. For a Newtonian fluid Chan-Leal's theory gives an equilibrium position of $s \approx 0.49$. It indicates that the equilibrium position of bubble shifts further towards the inner cylinder when shear thinning behavior is introduced ($s \sim 0.377 < 0.49$). This result is essentially in agreement with our experimental observations for shear thinning fluid (c.f. Fig.6.6). The predicted equilibrium position is different from the one in experiments and this is due to the different geometry chosen in simulations. Moreover, fig. 6.10 shows that the final equilibrium position does not depend on the shear rate. This is consistent with our experimental observations reported in figure 6.6 and also prediction of Chan-Leal's theory.

Furthermore, Fig. 6.11 shows the bubble trajectories for different bubble



Figure 6.10: Bubble migration trajectories in shear thinning fluid for R/d = 0.05 at different shear rates.

sizes and fixed shear rate. The final equilibrium position clearly moves away from the inner cylinder when bubble size is increased. On the other hand, the final equilibrium position for different bubble size tested in experiment is $s = 0.25 \pm 0.01$. Therefore, we do not see the effect of bubble size in experiments. Mason et al [26] have also measured the trajectory of droplets in shear thinning fluid with different size and did not see a considerable change in equilibrium position. They reported equilibrium position of $s \approx 0.43 \pm 0.05$ for R/d = 0.073 and $s \approx 0.453$ for R/d = 0.11. At this moment we do not have an explanation for the disagreement between experiments and simulations.

To rationalize the effect of shear-thinning on the final equilibrium position of the bubble, we consider two lateral forces acting on it. The first arises from wall repulsion, and is the same as in a Newtonian fluid that can be estimated from the Chan-Leal formula. The second is due to shearthinning. Specifically, we expect the inner side of the bubble to experience a higher shear rate and thus lower viscosity than the outer side. This radial asymmetry in the viscosity is probably the direct cause of an inward lateral force on the bubble, which shifts the bubble closer to the inner wall against the wall repulsion.



Figure 6.11: Bubble migration trajectories in shear thinning fluid for $\dot{\gamma} = 2.74s^{-1}$ at different shear rates.

We can estimate the net lateral force on the bubble from the computed bubble trajectory. To convert the lateral velocity of the bubble to a force, we borrow the following formula from a cylinder moving in a fluid bounded by a wall [36]:

$$F_l = \frac{4\pi\mu V_{mig}}{\left[\log\left(\frac{g+a}{R}\right) - \frac{a}{g}\right]},\tag{6.4}$$

where $a^2 = g^2 - R^2$. R and g are the bubble size and its distance from the wall. Fig. 6.12 compares this force experienced by a bubble sheared in shear thinning fluid with the one in Newtonian fluid. It appears that increasing of the shear rate does not change this force balance. But, increasing of the bubble size tends to empower the wall repulsive force. Consequently, the equilibrium position of the bubble shifts further away from the inner cylinder for larger bubbles.

Now we turn to the foam that has both shear-thinning and elasticity. We know that elasticity generates a net outward motion of bubble which opposes the inward motion of the bubble. This force is on the same order of magnitude as the shear thinning force but in the opposite direction. Therefore, if we combine these two effects, the net outcome is that the final



Figure 6.12: Calculated force for shear thinning fluid and Newtonian fluid vs dimensionless position on a bubble with in wide-gap Couette geometry. Newtonian force was evaluated using Chan-Leal formula.

equilibrium position should be in the range of equilibrium positions bounded by those for the purely shear-thinning and purely elastic fluids.

For migration experiments in foam the final position is between those expected from the two "rheologically pure" liquids. Although this force balance is a simplified version of what happens in real case, it is capable of rationalizing the experimental observations nonetheless.

In our discussion so far, we have tacitly taken the 2D foam of smaller bubbles as an effective continuum, a non-Newtonian fluid exhibiting shearthinning and normal stress difference. The conclusion of the above discussion is that as far as the bubble migration is concerned, the analogy seems to hold. This amazing fact is reminiscent of the use of the falling ball rheometer for measuring the viscosity of a suspension, made of particles comparable in size with that of the falling ball [4]. Nevertheless, the foam is a heterogeneous medium and its granularity manifests itself in certain ways. An example of this is the so-called nonlocal effect. Figure 6.2 shows the bulk shear rheology of the foam, measured from the torque on the inner cylinder. Alternatively we can measure the velocity profile of the foam using PIV [57], with typical profiles shown in Fig. 6.13. In the same figure we have plotted the velocity



Figure 6.13: Normalized velocity versus normalized radius. Symbols show the local measurements by using PIV and curves show the corresponding predictions using global measurements.

profile computed from the Herschel-Bulkley viscosity of the foam, and there is a disagreement between the global measurement (of viscosity) and the local measurement (of the v(r) profile). Katgert *et al.* [39] rationalized this discrepancy by a nonlocal effect in flowing foam, with clusters of bubbles moving cooperatively over a certain correlation length. This serves as a reminder of the subtle dynamics of sheared foam and of the limitations of the continuum analogy.

6.4 Conclusion

To conclude, we studied lateral migration of a bubble in sheared two-dimensional foam in a wide-gap Couette geometry. We reported two thresholds for shear rate and bubble size ratio. The final equilibrium position of a migrating bubble in wide-gap geometry differs from the narrow gap device. We showed that for a wide-gap geometry equilibrium positions shifts further towards the inner cylinder compared to a narrow-gap device. This shift was attributed to the non-Newtonian rheology of foam. It appears that the viscoelasticity of foam alters the migration behavior. We then understood the effect of shear thinning and elasticity on the migration of a bubble by conducting experiments on xanthan gum and Boger fluids separately. It is of interest that we could rationalize the migration results in foam by using the migration experiments in those aforementioned fluids. Foam also shows non-local effects in terms of rheology [39]. In this work we used the result of bulk rheology to justify the migration experiments, how about using local measurements? i.e. Can local measurements of rheology be used to understand migration in foam? This remains an open question that can be addressed in future studies.

Chapter 7

Conclusion and recommendations

The overarching theme of the research conducted in this thesis is to use the experiments to explore the structural evolution of two-dimensional foam undergoing flow. This is a necessary and significant step in studying the flow of foams in microscopic as well as macroscopic level. Up to now, most experiments on foams have focused on the structural changes in static or quasistatic states, and less attention has been given to the changes in structure in dynamical processes. There have been a limited number of experiments which investigated the dynamical change in foam structure. Those experiments are not in line with each other when they are put in one picture. For example, some experiments show that size based segregation happens in three dimensional foams in which larger bubbles tend to move away from the walls, but some other experiments in two-dimensional foams showed that large bubbles rather to move towards the wall. Therefore, knowledge in this area is limited and has lacked a firm scientific foundation. A well designed experiment with a simplified geometry fills a much needed role in this context.

Developing a new shear device with Couette co-axial cylinder geometry have allowed us to study the dynamics of two-dimensional foam thorough in simple shear flow. This device is coupled with three different cameras that allowed us to visualize the microstructure of the foam as foam undergoes shear simultaneously. The use of two-dimensional foam is preferred to threedimensional foam, since the latter is opaque and its microstructure is difficult to visualize.

We also used the PIV method to track and compute the velocity profile of foam across the gap and used this as additional tool to rationalize the experimental observations. In view of the outcome of the four projects, one can claim that the experiments have succeeded in identifying and clarifying the mechanisms underlying structural changes in foam. In addition, we have uncovered novel physics in sheared two-dimensional foams.

In the following, I will first summarize the key findings, and then reflect on their significance and limitations.

7.1 Summary of key findings

7.1.1 Coalescence of bubbles in sheared two-dimensional monodisperse foam

To thoroughly understand the mechanisms responsible for structural evolution in three-dimensional foams, we made the problem tractable by simplifying the sample to two-dimensional foam. This allowed us to simply visualize the foam structure at any point during experiments. We performed experiments in simple shear flow of two-dimensional foam in fairly high shear rates and observed appearance of large bubbles shortly after the start. Smaller bubbles have been coalescing to make larger bubbles only if a minimum threshold is passed. We did not observe breakup of bubbles in this case. Then, we explored the effect of different parameters including bubble size, viscosity of liquid and shear rate on the coalescence process. The minimum threshold for coalescence of bubbles in foam contradicts the conventional wisdom on coalescence in which coalescence occurs for gentler collisions. Then, we made an effort to rationalize the experimental observations using different theories that could possibly explain the results. Though none of the theories worked quantitatively, the most promising one was the model based on inertia of fluid. This theory can explain the minimum threshold for start of coalescence and the dependency of this shear rate to the bubble size, but overestimate the effect of viscosity.

Perhaps the most important finding of this study is that if we shear a two-dimensional foam fast enough, we would observe the coalescence of bubbles and therefore, formation of new large bubbles in foam. This is the first observation of its kind.

7.1.2 Cross stream-line migration of a single large bubble in monodisperse foams.

One of the potential mechanisms for structural evolution in foam is crossstream line migration of bubbles away from walls during the flow of foam. This phenomena has been extensively reported in flow of suspensions and emulsions. For foams, on the other hand, there have been relatively few studies on structural evolution in the literature, and they tend to contradict each other. Therefore, it is vital to understand the physics of the problem to resolve the apparent contradictions.

In this study we used a simple Couette co-axial cylinder and filled it with monodisperse foam. Then, we introduced a single bubble whose size was different than its surroundings. We reported cross-streamline migration for the cases in which bubble size ratio and shear rate were both above some thresholds. For those cases, bubble migrates from any initial position to the middle of the gap. Then, we modified a model based on perturbation theory to account for the migration in foam. The results of theory and experiments were in good agreement with each other. In addition to that we used a force balance between wall repulsion and capillary attraction to account for the presence of thresholds; again this simple force balance was in reasonable agreement with experimental observations. This was a key step towards understanding the mechanisms responsible for structural evolution in a confined foam flow.

7.1.3 Size-based segregation in sheared two-dimensional polydisperse foam

Following the study of single-bubble migration, the next step for us was to introduce more than one large bubble into a monodisperse foam and perform similar experiments as before in Couette co-axial cylinder geometry. When we have several large bubbles in a sea of smaller bubbles, they will interact with each other resulting in an effective diffusion. This diffusion tends to evenly distribute the large bubbles in the gap while the lateral migration does the opposite. The competition between these two mechanisms lead to distribution of large bubbles across the gap, which is peaked at the middle of the gap. There are also regions close to walls where no large bubble can be found. Then, we studied the effects of different parameters such as bubble size ratio, shear rate, area fraction of large bubbles and initial configuration of foam to better understand the process of size-segregation. Here, we again observed thresholds for migration similar to previous work. Beyond these thresholds, foam structure evolves to the state where large bubbles are mainly accumulated in the middle of the gap. Initial configuration of polydisperse foam does not seem to have any effect on the final equilibrium state as long as we are above the thresholds. Then, we used a model that accounts for both cross stream-line migration and diffusion due to interaction of large bubbles with each other. It turned out that model could predict the experimental observations to a good degree.

7.1.4 Effect of non-Newtonian rheology on bubble migration in sheared foam.

We have done experiments in a narrow gap Couette co-axial cylinder in which two-dimensional foam behaved essentially as a Newtonian fluid. Therefore, the non-Newtonian feature of the foam has not been manifested in previous experiments. One naturally wonders how does the non-Newtonian behavior of foam impact the structural evolution? To explore this question, we widen the gap of Couette co-axial cylinder to introduce a nonuniform shear rate profile within the gap. Two-dimensional foam in wide gap exhibits non-Newtonian behaviors including the shear thinning, yielding and elasticity. For simplicity we have focused on experiments in which foam is

fully yielded in the gap. Therefore, the remaining non-Newtonian features are shear thinning and elasticity. We made a monodisperse foam and placed it between two cylinders and then introduced one large bubble inside the sea of smaller ones. We again report the lateral migration for shear rates and bubble size ratio above some thresholds, similar to our previous results. The main difference between wide-gap experimental results and narrow gap is the final equilibrium position that the bubble attains. For narrow gap geometry this position is at the middle of the gap, while for the wide-gap geometry it is at the inner half closer to the inner cylinder. We also modified the perturbation theory of Chan-Leal to account for trajectory of bubbles. In this model we included the curvature of stream-lines, shear rate profile and elevated deformation of bubble. It turned out that model fails to explain the experiments. Therefore, there has to be another factor that leads to this deviation. We believe that this is due to the non-Newtonian features of the foam. To understand the process of migration in a viscoelastic fluid, one has to study the effect of shear thinning and elasticity separately. For foam this is impossible. Therefore, we designed some polymer solutions which mimic the shear thinning behavior and elasticity of foam. In addition to that, we used theoretical as well as simulation results to explain these two effects separately. Both experiments and calculations are pointing to the same direction that the shear thinning behavior pushes the bubble further towards the inner cylinder while the elasticity does the opposite. Therefore, for a foam which is viscoelastic fluid the final equilibrium position has to be in the middle of values for shear thinning and elastic fluids. This is consistent with the results of experiments in foam.

7.2 Significance and limitations

The insights gained from this research are potentially useful in two general ways. First, the novel phenomena that we have discovered in foam experiments may inspire further, more in-depth research in foam and flow of other multicomponent fluids. These experiments serve as the first window towards a better understanding of fundamental dynamics of foam as complex fluids. Second, This study provides potential guidelines for designing bubble structures in engineering processing. This understanding in turn would lead to products with better qualities.

For example, the final properties of some cosmetic products such as shaving cream is determined by their microstructure. It is shown in this thesis that the structure of a sheared foam tends to evolve, therefore, the quality of product would change. Hence, one can tune foam's properties by knowing its dynamics through the process of manufacturing. In another example, foam is used in process of enhanced oil recovery. Its efficiency can be improved by tuning its structure. To do so, one should identify the mechanisms responsible for structural evolution in foam and then try to tune them based on the requirements in each application.

The limitations of this research can be summarized as follows:

- 1. Two dimensionality. A real liquid foam has a complex 3D structure, but the opacity of three dimensional foam has forced us to study the 2D foam. With this simplification we might lose some basics physics which emanates from three dimensionality of foam. For instance, a bubble is surrounded by more bubbles in 3D foam than in a 2D one. This might change its deformation in the same flow field and therefore, its dynamics.
- 2. Limited range of bubble size. We could not test a very wide range of bubble size in the experiments with narrow gap Couette device due to some limitations. Foam structure has to be stable in the absence of dynamic flow field. In order to increase the stability of foam we needed to increase the viscosity of soap solution. This would result in bubbles with larger size. Moreover, we are bounded to put at least 10 bubbles across the gap of narrow Couette as a rule of thumb to assume the foam as a continuum fluid. Given the gap size, this gives us an upper bound for the bubble size.
- 3. Limited range of shear rate. For the Couette co-axial geometry we are bound to use a limited range of shear rate due to the presence of some complicating forces. If the shear rate is too high, centripetal forces come into play and contaminate the clear picture of lateral migration or size segregation in foam. The shear rate is also bounded from below by the need to ensure yielding throughout the entire domain. This avoids the unnecessary trouble of dealing with an uncertain yield surface in the experiments. Therefore, we had to make sure that we were operating within the range that the inertia of the fluid and also the yielding could not have affected the lateral migration and size segregation experiments.
- 4. Different geometry or flow field. Currently, our experimental observations are limited to Couette co-axial cylinder geometry. What about some other geometries such as channel flow or simple shear flow in parallel plate geometry? We can avoid the previous limitation for the shear rate range by investigating a different geometry.

7.3 Recommendation

We have taken the fist step to identify the main mechanisms responsible for structural evolution in polydisperse foam. One can carry out experiments to further detailed understanding of foam dynamics. In the following, I list some potential works that can be carried out in future.

1. Size-based segregation in wide-gap Couette co-axial geometry.

Following chapter 5, one can perform experiments on polydisperse foam and investigate the effect of nonuniform shear rate on size-based segregation. Then, possibly a model can be developed to describe the distribution of large bubbles among smaller ones similar to results reported in (chapter 5) for narrow gap system.

2. Correlation between Rheology and Size-based segregation in Couette co-axial geometry.

Another potential future works on polydisperse foam is to study the rheology of two-dimensional polydisperse foam in conjunction with the visualization techniques. We have studied the size-based segregation in polydisperse foam in chapter 5. Now, one can measure the rheological properties of two-dimensional polydisperse foam as its structure evolves. Would there be any correlation between size-based segregation and rheological properties of foam? And if yes, what is the underlying physics behind this correlation?

3. Complex flow fields and geometries.

There are also other factors that can be investigated. For instance, what is the effect of different flow fields on the aforementioned observations in chapter 4 and 5? Since foam processing necessarily involves complex flow fields that combine shear and elongation, and are spatially heterogeneous, it will be highly valuable to test foam flows in such complex geometries.

This summary makes it clear that there are myriad new problems that remain to be investigated in flows of foam. It is our hope that the findings of this thesis will inspire other researchers to contribute to this area of research, perhaps by bringing new tools and strategies to the as yet unresolved questions.

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