DYNAMICS OF RISING BUBBLES AND STRESS MEASUREMENTS THROUGH BIREFRINGENCE IN VISCOELASTIC LIQUIDS

by

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ABSTRACT

DYNAMICS OF RISING BUBBLES AND STRESS MEASUREMENTS THROUGH BIREFRINGENCE IN VISCOELASTIC LIQUIDS

Bubble dynamics in a viscoelastic liquid are studied to check the existence of a discontinuity in the terminal velocity that occurs at some critical bubble volume and cusp formation at the bubble tail and their relation with the stress around the bubble, which is measured through flow birefringence. A wide range of bubble volumes, $5-2000 \text{ mm}^3$ in 0.10-0.40 wt% polyacrylamide (PAAM) solutions is employed. The terminal velocity decreases with PAAM concentration and increases with bubble volume but plateaus. The plateau velocity is independent of PAAM concentration. No discontinuity in the terminal velocity is observed, which is supported by the bubble shape parameters of which circularity, the minor axis, and eccentricities are found to be concentration-dependent. Increasing the bubble volume changes the shape from spherical to prolate, teardrop with a cusp and cap-shapes. The critical bubble volume for the cusp formation is independent of the PAAM concentration. With the addition of 100 ppm surfactant SDS leads less distortion of the bubbles and cusp formation at smaller volumes. Large bubbles and dilute solutions cause bubbles wobbling. Increasing the PAAM and the surfactant concentrations shows an increase in the lateral direction motion. The retardance and the stretching angle around the bubble are measured for PAAM and hydroxypropyl cellulose (HPC) solution. For small bubbles, the stress is proportional to the concentration; whereas for large ones, bubble shapes become important for the stress. No significant effect of the temperature gradient on the bubble dynamics is observed. Weak convection patterns are seen through Schlieren method.

ÖZET

VİSKOELASTİK SIVILARDA YÜKSELEN KABARCIKLARIN DİNAMİKLERİ VE ÇİFTKIRILIM YÖNTEMİ İLE GERİLİM ÖLÇÜMLERİ

Bir viskoelastik sıvıdaki kabarcık dinamiği, bazı kritik kabarcık hacminde meydana gelen terminal hızdaki süreksizliğin varlığı ve kabarcık kuyruğundaki sivri uç oluşumu ve bunların, kabarcık etrafındaki gerilimle ilişkisi, akış çift kırılım yöntemi ile incelenmiştir. Ağırlıkça %0.10-0.40 poliakrilamid (PAAM) solüsyonları içinde çok geniş hacim aralığında, 5-2000 mm³, kabarcıklar kullanılmıştır. Terminal hız, PAAM konsantrasyonu ile azalmakta ve kabarcık hacmi ile artıp, plato yapmaktadır. Plato hızı, PAAM konsantrasyonundan bağımsızdır. Terminal hızda süreksizlik gözlemlenmemektedir ve bu şekil parametreleriyle desteklenmektedir; kabarcık daireselliği, küçük ekseni ve eksantrikliklerinin konsantrasyona bağlı olduğu bulunmuştur. Kabarcık hacminin arttırılması, şekli küreselden ovale, sivri uçlu gözyaşı damlasına ve kasket şekillerine değiştirir. Sivri uç oluşumu için kritik kabarcık hacmi, PAAM konsantrasyonundan bağımsızdır. 100 ppm sürfaktan, SDS ilavesi, küçük hacimlerde kabarcıkların daha az bozulmasına ve sivri uç oluşumuna yol açar. Büyük kabarcıklar ve seyreltik çözeltiler kabarcıkların yalpalayarak yükselmesine neden olmaktadır. Polimer ve yüzey aktif madde konsantrasyonlarının artması, yanal yön hareketinde bir artışa yol açmaktadır. PAAM ve hidroksipropil selüloz (HPC) çözeltisi içinde kabarcık etrafındaki retardasyon ve esneme açısı ölçülmüştür. Küçük kabarcıklar için gerilim konsantrasyonla orantılıdır; büyükler için ise kabarcık şekilleri önem kazanır. Sıcaklık gradyanının kabarcık dinamiği üzerinde önemli bir etkisi gözlenmemiştir. Schlieren yöntemi ile zayıf konveksiyon desenleri görülmüştür.

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LIST OF SYMBOLS

a	Viscosity constant of Carreau-Yasuda model
A	Cross section area of the bubble
Ar	Archimedes number
Bo	Bond number
Ca	Capillary number
C_D	Drag coefficient
d	Equivalent diameter
dc	Distance between cone and plate
De	Deborah number
E1	Eccentricity-1
E2	Eccentricity-2
Eo	Eötvos number
F	Force acts along the edge of the length
g	Gravitational acceleration
G'	Storage modulus
G''	Loss modulus
Ga	Gallilei number
Н	Optical length
h	Height of the truncated cone
Ι	Intensity of light
I_0	Intensity of the incident light beam
K	Consistency index
ℓ	Length of the edge
Mo	Morton number
n	Power-law index
r	Radius of the lower section of the bubble
R	Radius of the upper section of the bubble
r_c	Cone radius of the rheometer

r_r	Ring radius
Re	Reynolds number
u_B	Terminal velocity
V	Bubble volume
V_i	Volume of i th cone
We	Weber number
β	Empirical shape parameter
γ	Shear strain
$\dot{\gamma}$	Shear rate
δ	Retardance
η	Viscosity
η_0	Zero shear rate viscosity
η_a	Apparent viscosity
η_{∞}	Viscosity as the shear strain rate goes to infinity
heta	Angle between cone and plate
λ	Relaxation time
λ_i	Wavelength of the incident light
П	Correlation parameter
ρ	Density
$ ho_\ell$	Density of the Liquid
$ ho_g$	Density of the gas bubble
σ	Surface tension
τ	Shear stress
χ	Stretching angle of the polymer

LIST OF ACRONYMS/ABBREVIATIONS

CMC	Carboxymethylcellulose
FPS	Frame per second
HPC	Hydroxypropyl cellulose
LVE	Linear viscoelastic region
PAAM	Polyacrylamide
PIV	Particle imaging velocimetry
SDS	Sodium dodecyl sulfate

1. INTRODUCTION

For hundreds of years, multi-phase flows have been a topic of interest due to its wide application areas such as chemical, petrochemical, food, nuclear processes etc. Some of the important applications can be gas and oil pipelines, nuclear reactor, inkjet printing and chemical reactors etc [1–3]. In this study, we are interested in the two-phase flow studies; in particular bubbles rising in viscoelastic liquids.

An important part of the literature deals with the gas bubbles in a Newtonian medium. While most gases and substances exhibit Newtonian behaviour, there has been a growing interest in recent years to understand the characteristics of the twophase flows employing non-Newtonian liquids as they are found both in nature (gums, honey, biological fluids such as blood, mucus and synovial fluids) and in industrial processes (high molecular weight polymers, emulsions, plastics and some kind of dispersions). Due to the complex nature of the non-Newtonian liquids, the study of bubble motion in non-Newtonian liquids has been a challenging subject. There exists different types of non-Newtonian liquids such as Bingham plastics, Bingham pseudoplastics, shear-thinning and shear-thickening; and only shear-thinning liquids are employed in this study. Additionally, the viscoelasticity is also studied [4–6].

Bubbles are different than the rigid particles due to their mobile surfaces and their surface deformability. The bubble shape is deformed depending on the stress on it during its motion. Bubbles can exhibit different types of shapes such as spherical, prolate teardrop, oblate cusped-like and spherical cap [7]. Their motion in the non-Newtonian solutions is also different from the bubble motion in Newtonian solutions. In the literature, it is shown that it is difficult to predict the bubble velocity and shape while rising in a shear-thinning and viscoelastic liquid. Some studies reported a sudden increase in the bubble velocity up 10 fold, in polyacrylamide (PAAM) solutions [8–11]; while a jump was not reported in some other polymer solutions [7, 8, 12, 13]. The reasons attributed to the presence of the velocity jump are the cusp formed at the tip of the bubble and the downwards velocity field, i.e., the negative-wake at the tip of this cusp [13]. The volume where the jump occurs is called the critical volume. The jump is observed at the transition from the Stokes to the Hadamard-Rybczynski regime for some studies [9]. However, there are studies where a jump is not observed even though they report the cusp-like shape and the negative-wake [11, 13–20]. The dimensionless numbers such as Reynolds, Re, Bond, Bo, capillary, Ca and Morton, Mo are used to understand the system and find the critical bubble volume where the system shows a different behaviour. The bubble shape is also an important parameter as the jump is always observed at the cusp-like shape. The bubble deformation is used to classify the bubbles according to their shapes. Particle imaging velocimetry (PIV) is a technique that is preferred to visualize the velocity field [15–17,21,22]. Also the birefringence, an optical method, is used to visualize the stress around the rising bubble [17–20]. The reader is referred to Clift *et al.* (1978) and Chhabra (2006) for a detailed study that considers the behaviour of particles, drops and bubbles in non-Newtonian liquids [1,7].

It can be seen that there are various unanswered questions in bubbles rising in non-Newtonian fluid systems. Even tough, the previous studied had several hypotheses, there is no certain answer why some of the studies observed the jump and the others did not. In this work, the aim is to find answers to these questions or evidences to understand the bubble dynamics better.

Based on the literature, it is hypothesized that there can be a critical bubble volume where the bubble shape, the average terminal velocity of the bubble, the polymer concentration and the stress around the rising bubble are directly correlated with each other. This study aims to show how the introduced parameters change in a system for a broad range of bubble volume, surfactant and polymer concentrations. It is expected that the findings of this study will contribute to the literature by showing which parameters are important and needed to be studied to understand the behaviour of bubble in terms of shape, velocity and stress, while rising in viscoelastic liquids.

For this purpose, the dynamics of a bubble rising in viscoelastic and shear-

thinning PAAM solutions with different concentrations are studied using multiple deformation parameters and dimensionless numbers. To illuminate this uncharted area, we examined the stress around the bubble as it rises using the birefringence method. In particular, no study, to our knowledge, has considered to quantify the stress to search if there is a relationship between the stress and the bubble dynamics using the birefringence method. Moreover, the effect of the surfactant on the bubble dynamics is studied. Finally, the bubble dynamics is examined in the presence of a temperature gradient.

A literature survey is presented in Section 2. In Section 3, the information about the materials and the rheological characterizations of the solutions are given. The experimental setups and the methods used in the experiments are presented in Section 4. The parameters that are measured during the experiments are introduced in Section 5. Image processing and the techniques used to analyze the experiments are studied in Section 6. In Section 7, the results are presented and discussed. Conclusions and recommendations are given in Section 8. Uncertainty analyses are presented in Appendix A for the experimental measurements and the calculations. Finally, the permissions for the figures taken from the literature are given in Appendix B.

2. LITERATURE SURVEY

In this section, a literature survey on the dynamics of a rising bubble is presented. In Section 2.1, non-Newtonian liquids and the rheological properties are introduced, and the constitutive equations used in this study are given. The dynamics of a bubble rising in viscoelastic solutions are studied in Section 2.2 in detail. This Section also includes the studies related to the injection period, the use of surfactant, the bubble trajectory and the birefringence method.

2.1. Non-Newtonian Liquids and Rheology

It is known from the two-plate model that when one of the plates is sheared such that it moves at constant speed u (see Figure 2.1), then there is a relationship between the applied shear stress, τ and the shear rate, $\dot{\gamma}$, as

$$\eta_a = \frac{\tau}{\dot{\gamma}} \tag{2.1}$$

where η_a is the apparent viscosity.



Figure 2.1. Two plates model for the viscosity.

For Newtonian fluids, the viscosity is independent of the shear rate, and is shown as η . For non-Newtonian fluids, the fluid viscosity changes with shear rate. Although simple gases and liquids show Newtonian behaviour, the behavior of complex systems such as emulsions, suspensions, solutions containing long molecules or fluids with high molecular weight is more complex [23].

Non-Newtonian fluids are classified as time-dependent, e.g., thixotropic and rheopectic and time-independent, e.g., Bingham plastic, Bingham pseudoplastic, dilatant, pseudoplastic, shown in Figure 2.2 [4–6,24].



Figure 2.2. Time-independent fluids.

In this study, we are only interested in pseudoplastic (shear-thinning) fluids. The viscosity of this type of fluid decreases with shear rate (Figure 2.3). Most non-Newtonian fluids are in this class and include polymer solutions or melts, greases, starch suspensions, biological fluids, detergents, dispersion media of some drugs and dyes [25, 26]. The reader is referred to [25] for more detail about other types of non-Newtonian fluids.



Figure 2.3. Flow curves for shear-thinning fluids.

2.1.1. Non-Newtonian Rheology Models

The power-law, Carreau-Yasuda, Herschel-Bulkley, Cross and Casson models are the most widely used rheological models. In this dissertation, only the power-law and the Carreau-Yasuda models are employed; therefore, only these two models are presented here.

<u>2.1.1.1. Power-Law Model.</u> The power-law model is widely used to characterize semisolid materials. In addition to being a well-known and useful model, the power law model is very important as it is applicable in the $10^1 - 10^4$ s⁻¹ shear rate range where many commercial rheometers operate. The disadvantage of the power-law model is that materials exhibiting shear-thinning and shear-thickening behaviors cannot be identified at low and high shear rates [24, 27, 28].

The power-law model is a two-parameter model where The flow consistency index, K (Pa.sⁿ) is the shear stress at 1 s⁻¹ shear rate, is given as follows:

$$\eta_a = K \dot{\gamma}^{n-1}. \tag{2.2}$$

The power term n, which is the flow behaviour index, is unitless. In order for the experimental data to be compatible with this model, K > 0 and 0 < n < 1 for the shear-thinning fluids. For the special case of the Newtonian fluids where n is equal to 1, the consistency index K is equal to the viscosity of the fluid. The fluid exhibits shear-thickening behaviour when n is greater than 1.

When the logarithms of both sides of Eq. 2.2 are taken, the following equation is obtained,

$$log(\eta_a) = log(K) + (n-1)log(\dot{\gamma}) \tag{2.3}$$

where K and n parameters are determined from the intercept and the slope of $log\eta_a$ vs. $log\dot{\gamma}$ plot.

Shear-thinning fluids that are interest of this study can be described through the power-law model; but exhibit Newtonian behavior at extreme shear rates. Therefore, the viscosity versus the shear rate graph can be divided into three regions:

- (i) low shear rates: the viscosity is constant at, η_0 .
- (ii) medium shear rates: the viscosity changes with the shear rate where the powerlaw model is applied.
- (iii) high shear rates: the viscosity is constant at its infinity value, η_{∞} .

The described behaviour including the region where the power-law model is applied, can be seen in Figure 2.4. The Carreau-Yasuda model can cover the entire forementioned behaviour and is explained in Section 2.1.1.2.

<u>2.1.1.2. Carreau-Yasuda Model.</u> It is a model that represents the polymer flow behavior in a wider range of shear rate than the power-law model. Unlike the power-law model, it is a model in which the η_0 and η_∞ parts can also be represented (Figure 2.4).



Figure 2.4. The representative behaviour of the viscosity of shear thinning fluids.

The Carreau-Yasuda model is given as follows,

$$\eta_a = \eta_\infty + (\eta_0 - \eta_\infty) (1 + (\lambda \dot{\gamma})^a)^{\frac{n-1}{a}}$$
(2.4)

where, λ is the relaxation time and used to determine the transition point from the zero shear rate zone to the shear-thinning behavior. The constant, *a* determines the shape of the transition between the constant viscosity region and the power-law region. If *a* is smaller than 1, the transition zone becomes longer. If *a* is greater than 1, the transition will be instantaneous. Here, *n* represents the flow behaviour index again. The Carreau-Yasuda model is very flexible as it is controlled by five different parameters (η_0 , η_{∞} , λ , *a* and *n*). Therefore, it is the most widely used shear viscosity model [4, 25, 28, 29].

2.1.2. Viscoelasticity

In this study, we are particularly interested in viscoelastic fluids. Viscoelasticity gives information about the elastic behavior of viscolelastic solids and the flow behavior of liquids. In order to understand the viscoelasticity better, it is necessary to know what the elastic and viscous behavior is. When an external force is applied to an object, the substance changes its shape and when this force is removed, the object can return to its original state. This behavior is called as the elastic behavior. If an external force is applied to the object and the object shows delayed deformation, it exhibits viscous behavior. The difference between the delayed strain and the stress is called the phase angle. Viscoelastic behavior depends on the relationship between the elasticity and the viscosity of the material with respect to this phase angle. The stress/strain ratio obtained in rheological studies gives the modulus of the material. While examining the elastic structure of the material, the storage, G' modulus is taken into account, while the loss, G" modulus is taken into account to examine the viscous structure of the material. Viscoelasticity is studied in detail through the experimental rheological characterization in Section 3.2.

2.2. Bubble Dynamics in Viscoelastic Liquids

The shape and the velocity of a rising bubble depend on its size and the physical properties of the liquid, e.g., the viscosity, the density and the surface tension. The liquid-gas two phase flow is often employed in many industrial sectors, including energy generation, chemical, food, biochemical, and waste water disposal [2]. Due to the complex rheological properties of the non-Newtonian fluids such as shear-thinning, viscoelasticity, etc., the dynamics of free-rising bubbles in non-Newtonian fluids are very complex.

One of the earliest studies about the bubble motion in non-Newtonian fluids is presented by Astarita and Apuzzo (1965) [8]. They experimentally studied the terminal velocities of bubbles rising in different non-Newtonian fluids (Carbopol, carboxymethylcellulose (CMC), ET497 and J-100), which differ by viscosity and elasticity. They reported a discontinuity upto 6 folds at the bubble rise velocity at a specific bubble volume for highly elastic solutions (ET497 and J-100). This discontinuity was called a jump where the velocity value increases abruptly above a critical volume. In this work, the jump was attributed to the viscoelasticity. Later, Leal *et al.* (1971) experimentally and theoretically investigated the discontinuity using PAAM solutions (Separan AP30) [9]. Their hypothesis was that a change in interfacial conditions due to the no-slip boundary condition to free shear caused a 5-10 fold of discontinuity at the velocity. They tested this hypothesis by comparing the terminal rise velocities of bubbles and glass spheres with the same radius. They observed no jump at the velocity of the glass sphere; but a jump in the velocity of bubble. Therefore, it was believed that the discontinuity was caused by the transition from the Stokes to Hadamard regime [9]. Acharya et al. (1977) also observed the discontinuity for CMC and PAAM solutions and linked the reason for this to the mobile surface of the bubbles [10] and the polymer molecules acting like a surfactant on the bubble surface. However, they also added the size of the jump was less than the previous studies [8,9]. They reported that the inertial stresses had an effect on the bubble deformation where the fluid properties of the solutions, i.e. the viscoelasticity and the surface tension are kept constant; and the elastic properties was turning bubbles into prolate and oblate shape for low and high Reynolds number, Re (the ratio of inertial forces to viscous forces), respectively. In line with the study of Astarita and Apuzzo (1965) [8], Dekee and Chhabra (1986) confirmed the absence of the discontinuity for CMC solutions [12]; and the small size of the jump observed by Acharya *et al.* (1977) can be explained by the low elasticity of CMC solutions [10]. However, Dekee et al. (1986, 1988) [12,14] did not observe the discontinuity for the PAAM solution (Separan AP30) and had a discrepancy with Leal *et al.* (1971) [9].

Hassager (1979) did not report any discontinuity at velocity using laser Doppler anemometry for the PAAM-glycerol mixtures but showed the presence of the velocity directed downwards at the tip of the air bubble and having a tip similar to the knife edge also known as *cusp* [13]. Hassager (1979) called this downwards velocity *negative-wake* (zone 3 in Figure 2.5) and showed that bubbles had cusp (shown from two different angles in Figure 2.6), i.e. the sharp pointy tip at the tail above a critical bubble volume in the presence of the negative-wake. They observed that the tip was becoming weaker then evolving into a rounded shape as the bubble volume decreases. Coutanceau and Hajjam (1982) also studied the effects of the viscoelasticity and the shear-thinning
property on the bubble dynamics in different viscoelastic solutions [11]. They made a comparison between viscoelastic (PAAM Separan AP30, polyox and CMC) and Newtonian solutions with the same density and the viscosity. They concluded that the elasticity of a solution was the reason for the formation of the negative-wake; therefore, the increase in the bubble velocity and the cusp formation. They also showed that the velocity was reaching to a limit with increasing elasticity. However, a limit did not exist with the increasing shear-thinning effect [11]. Using the same polymer solutions with [13] and [11], Dekee *et al.* (1988) investigated the transition of the bubble shape as a function of the bubble volume and the polymer concentration in detail; and obtained four different bubble shapes, i.e. spherical, prolate, teardrop, oblate cusped and spherical cap with increasing volume [14].



Figure 2.5. The representation of the flow field and the negative-wake (zone 3) around a bubble rising in a viscoelastic and shear-thinnning solution (reprinted with permission from [17]).

In later works, the study of the bubble shape and the negative-wake became more important to understand and explain the bubble rise velocity in viscoelastic and shear-thinning liquids due to the disagreement in the literature. The presence of the discontinuity at the bubble velocity was attributed to the viscoelasticity of the solutions. The negative-wake , hence, the cusp, were also explained through the viscoelasticity. However, there were also cases where there was a cusp formation but no discontinuity in viscoelastic solutions. For example, Dekee *et al.* (1988) clearly showed the cusp formation for a rising bubble while reporting no discontinuity at the velocity [14]. Therefore, it is necessary to inform the reader about this uncertainty and the conflict at this point. The conflict has continued in the literature until today, and in this section we are providing the results of the related studies while discussing their evidences and explanations.



Figure 2.6. A bubble with a cusp at its tip (a) rising in a mixture of PAAM-glycerol; the perpendicular view of same the bubble (b) (reprinted with permission from [13]).

Along with the previous works, Miyahara and Yamanaka (1993) reported no discontinuity for CMC solutions and showed that for the spherical bubbles at low Re, the experimental drag coefficient, C_D is in an agreement with the Hadamard-Rybczynski type of drag coefficient, C_D . The experimental C_D deviates from the Hadamard-Rybczynski region as a cusp forms at higher Re [30]. Dewsbury *et al.* (1999) compared the rise of solid spheres to the rising bubble in CMC solution [31], and showed that the solid particles with the same volume and shape (bubble cap) as the bubbles, shared the same C_D curve for Re > 135. However, the bubbles fitted the Stokes model C_D for Re < 5 where the solid particles did not. The findings of Margaritis *et al.* (1999) about C_D for Re < 1 in viscoelastic and shear-thinning polysaccharide solutions are also in an agreement with Miyahara and Yamanaka (1993) [30]. They showed that C_D reaches a value of 0.95 with increasing Re; and the terminal rise velocity does not show any discontinuity while reaching a plateau for the increasing bubble volume. Same observations were done by Amirnia et al. (2013) for CMC and xantham gum solutions [32]. The validity of the Hadamard-Rybczynski type C_D was also proved for other viscoelastic and shear-thinning solutions such as xanthan gum solution [33]. Apart from these works, Zhang *et al.* (2008) developed an empirical C_D formula [34] and demonstrated that viscoelastic and shear-thinning solutions behave different than the solutions that show only shear-thinning behavior. Therefore, they proposed a modified Re that includes the elongation time, λ to calculate C_D for the accelerating bubble motion [34]. Wenyuan et al. (2010) also developed an empirical correlation for C_D that is a function of Re, Archimedes number, Ar (the ratio of the gravitational forces to the viscous forces) and Eötvos number, Eo (the gravitational forces compared to the surface tension forces) [35]. This type of C_D is different than the previously mentioned ones by including the bubble shape parameter, i.e. the aspect ratio. The C_D proposed by Wenyuan *et al.* (2010) was found to be in a good agreement with the C_D formulas developed by Dewsbury *et al.* (1999) and Zhang *et al.* (2008) [31, 34].

Wenyuan *et al.* (2010) indicated that for a given Re and Eo, the bubble shape changes for different type of polymers for a given [35]. Similar to the work of Wenyuan *et al.* (2010) [35], Li *et al.* (2012) developed an empirical C_D . However, they included the flow behaviour index, n of power-law model instead of λ . The proposed C_D covers a wider range of Re (0.05–300). They also provided a shape diagram to show the transition of the bubble shapes. The literature has been in an agreement that the rising bubble obtains a spherical shape due to the surface tension forces at very low Re. But a transition occurs in the shape depending upon the bubble volume and the physical properties of the surrounding phase. An early work by Grace (1973) proposed a phase diagram, *Grace-map* in terms of Re, *Eo* and Morton number, *Mo* where one can successfully predict the shape of the bubble rising in non-Newtonian liquids [1,36]. The Grace-map has been used widely [35,37]. One of the recent studies who also used the Grace-map, Xu *et al.* (2019) proposed two new shape parameters to characterize the shape of the rising bubble for xanthan gum and PAAM solutions [37]. They quantified the cusp formation through one of the proposed parameters.

Another important dimensionless number to study bubble dynamics is capillary number, Ca, which shows the ratio of the viscous to the surface tension forces. For polyox solutions, Liu *et al.* (1995) showed that the cusp formation and discontinuity occur up to 10 fold when the Ca is around unity, 1 [38]. They believed that the discontinuity was due to the cusp formation near the critical Ca value. However, when Wagner *et al.* (2000) did simulations for a similar system, they did not observe any jump due to the cusp formation [39]. Besides, Rodrigue *et al.* (1998) also did not observe any jump for low concentrations of PAAM (Separan AP273) solutions when Ca was around 1 [40] in the presence of a cusp. However, for higher, the critical Ca is around 1.

Soto *et al.* (2006) showed that the shear rates where they observed the discontinuities for different concentrations of the HASE solutions, corresponded to the appearance of normal stresses in the rheology analysis [41]. This is the first study that mentions about a critical shear rate value. They proposed that the cause of the discontinuity is the balance between elastic and surface tension forces. Pillapakam and Singh (2006) also considered the elastic and the surface tension forces. They indicated that the elastic forces being much larger than the surface tension forces causes the cusp formation [42]. However, unlike Soto *et al.* (2006) [41], they did not report any discontinuity [42]. Soto *et al.* (2006) also found out that the critical volume depends on the polymer concentration and decreases with it. Whilst this result aligned with a prior research of Herrera *et al.* (2003) [15], it had a conflict with Rodrigue and Dekee (1999) [43] as they said the critical volume is independent of the concentration. Pilz and Brenn (2007) considered both of these forces and experimentally obtained a universal correlation parameter to predict the presence/absence of the discontinuity [44]. This parameter, II was a function of λ , the gravitational acceleration, g, the density of the liquid, ρ and the surface tension, σ ; and defined as $\lambda(g^3\rho)^{1/4}$. The discontinuity is observed for different polymeric solutions, when Π was higher than 9.8938. No discontinuity is observed at low polymer concentrations and a cusp at the tip of the bubble does not ensure the presence of the discontinuity [44]. This universal correlation parameter, Π can be the most definite criteria to predict the behaviour of the bubble rise velocity. Hassan *et al.* (2010) investigated the effects of the viscous, inertia and surface tension forces on a bubble rising in xanthan gum solution through *Re* and Weber number, *We* (the ratio of the inertial to the surface tension forces for curved surfaces) [45]. Surface tension forces were dominant for small bubbles whereas both of inertia and surface tensions were effective for medium range of bubble volume. However, the inertia is dominant for larger bubbles. Viscous forces had a small effect for 5 < Re < 200 [45].

There are also some numerical work with various constitutive equations, considering bubble dynamics in viscoelastic liquids. Pillapakam and Singh (2001) used finite-element method (DNS) to show that bubbles have cusps after some critical values of Ca and Deborah number, De (represents the fluidity) [42]. The stability analysis of You *et al.* (2009) were in an agreement with Pillapakam and Singh (2001) as they also indicated the presence of a critical bubble volume and Ca for a specific De to form a cusp-like shape [46]. These studies support the findings of Wagner *et al.* (2000) and Malaga and Rallisonn (2007) who employed Lattice Boltzman and FENE models, respectively; and did not detect any discontinuity [39, 47]. In a later work, Lind and Phillips (2010) employed the boundary element method [48]. They imposed the irrotational flow assumption to prevent the negative-wake formation at the bubble's cusp-like tail. The results showed no discontinuity at rise velocity. Thus, Lind and Phillips (2010) deducted that the cusp shape is solely not enough, and a negative-wake is a prerequisite for the discontinuity [48].

Tripathi *et al.* (2015) executed unsteady simulations for a bubble rising in a Herschel-Bulkley liquid where they showed that the bubble did not reach the steady state for weak-surface tension cases [49]. The numerical simulations of Premlata *et al.* (2017) showed that as the shear-thinning property of the liquid increases, the bubble's shape deforms less and the rise velocity increases in the low Eo and Gallilei number, Ga (the ratio of gravitational to the viscous forces) regime [50]. In a very recent work, Yuan *et al.* (2020) employed rheoInterFoam solver that is developed for viscoelastic fluids and the solver employs the finite volume method [51]. They confirmed the findings of Premlata *et al.* (2017) about the low Eo and Ga regime.

In the literature, the particle image velocimetry (PIV) method was employed to examine the flow field around a rising bubble. Funfschilling and Li (2001, 2006) did not report any discontinuity for both CMC and PAAM (AN 913 SH and AN 934 SH) solutions while proving the presence of the cusp and the negative-awake (see Figure 2.5) using the PIV method [17, 21]. In a later work, Böhm *et al.* (2016) improved the flow field around the bubble using PIV [22] by showing that there actually exists 5 different flow zones (Figure 2.7) around a cusp-like shaped bubble instead of 3 zones (Figure 2.5) reported by Funfschilling *et al.* (2006) [17, 22]. The findings of Li *et al.* (2012) was in an agreement with Funfschilling *et al.* (2006) [21] for CMC solutions while showing that the viscosity distribution around the rising bubble using PIV. A high and low viscosity regions were observed for the center of the negative-wake and around it, respectively.

On the other hand, Herrera-Velarde *et al.* (2003) who also used the PIV method, showed that the discontinuity occurred when the negative-wake forms in the PAAM solutions (PAAM Separan AP30) [15]. They also added that the critical bubble volume where the jump occurs, decreased while the magnitude of the jump increases with the increasing polymer concentration. Similarly, Kemiha *et al.* (2006) employed PIV for PAAM solutions. They experimentally proved that the cause of the negative-wake was not due to the interface deformation by comparing a solid particle and a bubble [16]. As mentioned earlier, Dewsbury *et al.* (1999) also employed both solid and deformable subjects. Even tough, the rigid sphere and the bubble showed a discrepancy at C_D for low Re [31, 52], Kemiha *et al.* (2006) demonstrated the existence of a negative-wake for both, rigid and deformable surfaces [16].



Figure 2.7. The improved flow field and the negative-wake around a bubble rising in a viscoelastic and shear-thinnning solution (reprinted with permission from [22]).

Even tough it did not employ a shear-thinning liquid as the rest of literature given in this section, it is worth discussing the finding of Vélez-Cordero *et al.* (2012) here [53]. This study employed a Boger-type fluid that is an elastic liquid with a constant viscosity and independent of the shear rate. The purpose was to study the bubble rise velocity where only elastic forces were effective. A discontinuity was obtained at the bubble velocity; however, no negative-wake was observed. This result is particularly interesting as it shows that a negative-wake does not necessarily exist in the presence of a discontinuity [53]. They also mentioned that this critical bubble volume is not observed at Ca = 1 unlike Liu *et al.* (1995) and Rodrigue *et al.* (1998) [38,40].

2.2.1. Effect of the Injection Period on the Bubble Rise

An important parameter that has an effect on the bubble rise velocity is the bubble injection period. In an early study, Carreau *et al.* (1974) proved the effect of the period on the bubbles rising in CMC and PAAM solutions. Later, Dekee and Chhabra (1986) [12] stated that they did not observe any significant effect on the velocity and the bubble shape; and they attributed the dependency on the period reported by Carreau *et al.* (1974) to the orientations of the molecular chains of ionic polymers with high molecular weight [54]. Li *et al.* (1997a, 1997b, 1998) believed that the accumulated residual stress left behind a bubble needs to be considered due to its effect on the drag. Their observations showed that accumulated stress causes an increase in the rise velocity [55–57]. Li *et al.* (2004) showed that the injection period for a constant bubble volume becomes more important as the polymer concentration increases [19]. They reported a period upto 12 s is necessary.

In a later work, Funfschilling and Li (2006) compared the injection period for glycerol (Newtonian), CMC (similar viscosity with glycerol, slight shear-thinning) and PAAM (high viscosity and highly shear-thinning) [21]. Glycerol and CMC required an injection period higher than 0.3 s. However, 60 s of injection period is required for the PAAM solution. A lower injection period causes an increase in the bubble velocity for the PAAM solution as the bubble is affected by the wake of the previous bubble. It seems like the bubble shape does not change with the injection period for the CMC solution. However, a critical bubble volume exists for the PAAM solution where the injection period starts showing its influence [21]. Some other works preferred an injection period of 300 s [15,41,58].

2.2.2. Effect of Surfactant on the Bubble Rise

The bubble motion is also affected by the impurities that can be found on the surface of the bubbles. Small concentrations of surfactants can drastically alter the behaviour of a bubble. Clift *et al.* (1978) showed that the no-slip surface exists in the presence of the surfactant. The bubble rise velocity decreases with the addition of the surfactant into the solution due to the Marangoni effect where the surfactant concentration is not uniform along the surface of the bubble [59]. Surfactant changes the surface tension and the tangential shear stress occurs on the bubble surface. The presence of the surfactant in the medium also changes lateral movement of the bubble.

The bubble velocity decreases as the surfactant concentration increases [43,58,60] until the C_D value reaches the C_D of a rigid sphere. Karamanev (1994) stated that the bubble acts like a rigid sphere if the bubble surface is contaminated enough with the surfactant [61,62]. Cuenot *et al.* numerically showed that for a spherical bubble, the bubble velocity decreases, therefore C_D increases for Re < 100 [63]. Tzounakos *et al.* (2004) experimentally showed that the surface mobility does not have any influence on the bubble shape in the presence of the surfactant whereas the rise velocity is strongly depended on the surfactant concentration [60]. However, Li *et al.* (2018) also added that the rise velocity becomes independent of the surfactant concentration after a critical bubble volume [62]. It is observed that C_D behavior changes from Stokes to Hadamard-Rybczynski regime for a given Re range. [60].

Nalajala *et al.* (2014) simulated the bubble rise and the drag forces in a power-law liquid using ANSYS Fluent [64]. Their results proved the existence of a recirculating wake at the tip of the bubble above a critical Re and surfactant concentration. The wake grows with increasing shear-thinning property. They showed that the C_D increases as the liquid becomes more shear-thinning below a critical Re [64]. In a very recent work, Ahmed *et al.* (2020) studied the effect of soluble surfactant and the viscoelasticity on the bubbly flow where they employed FENE-P viscoelastic model and the front-track method [65]. They showed that when the surfactant concentration was high enough, the bubbles travel to the core region of the channel in the lateral direction. Marangoni stresses are cancelled out by the viscoelastic stresses. A bubble-wall layer forms due to the combination elastic, inertial and Marangoni forces [65].

2.2.3. The Bubble Trajectory

When a bubble rises in a liquid, it tends to have an axi-asymmetric trajectory such as zigzag or spiral, depending on the physical characteristics of the liquid. Hassan *et al.* (2008) showed that the large bubbles move spirally in xanthan gum solution while the smaller bubbles exhibit less horizontal move and do a zigzag motion at low Re and We. [33, 45]. It is found that the spiral movement of a large bubble is bigger for the bubbles rising in massecuites [45], a shear-thinning sugar solution with crystal suspension, than a solution without crystals [33].

Hassan *et al.* (2010) hypothesized that the zigzag motion is due to an instability caused by the vorticity created by the wake of the bubble [45]. Böhm *et al.*(2014) showed that the ellipsoidal shaped bubbles followed a rectilinear path in xantham gum solution [66]. Kumar *et al.* (2015) tested both shear-thinning and shear-thickening solutions [67]. They found that the bubble path does not depend on the shear-thinning or thickening property and is rectilinear [67]. Later, Xu *et al.* (2017) proved that the bubble trajectory depends on the bubble shape deformation in CMC solutions [37]. The rise of the bubble stabilizes as the polymer concentration increases. The oscillation of the bubble increases with the nozzle diameter [37].

2.3. The Birefringence Method for the Bubble Rise Dynamics

Some materials show anisotropic properties. The elongated shape of the molecules causes the anisotropic behavior of the liquid crystals. Some of the liquid crystals are in a cholesteric phase where the molecules are directionally oriented and stacked in a helical pattern [68]. These materials exhibit a property called birefringence, which is observed only in the presence of stress. Its magnitude of the refractive indices at each point in the material is directly related to the state of the stress at that point.

When a ray of light passes through a birefringent material, it becomes polarized, splits into two components (ordinary and extraordinary beams), and gets resolved along with the two principal stress directions. Each of these components experiences different refractive indices. The difference in the refractive indices leads to a relative phase difference between the two-component waves, which is usually called phase retardation. The magnitude of the relative retardation is given by the stress-optical law [69] which is explained in Section 5.2 in detail.

As mentioned before, PIV method can be used to visualize the flow field. As

an alternative, the birefringence can be employed to visualize what occurs around a rising bubble. The stress around the bubble can be quantified through the retardance and the stretching angle of the polymer through the birefringence method. In multiple works, Frank and his colleagues showed the stress distribution around the bubble rising in PAAM mixed with hydroxypropylcellulose (HPC) solution using the birefringence method [17–20]. HPC was added into the PAAM solution to add birefringence property to the solution. They did not quantify the stress. They visually showed the butterflyshaped stress distribution and the negative-wake around the bubble. They also did not report any discontinuity at the rise velocity for HPC added PAAM (AN 905 SH and AN 913 SH) solutions. Oba *et al.* developed polarization imaging method; and studied the birefringence and extinction angle distributions [70]. They showed their work is applicable to image the non-uniform flow in wormlike micelllar solutions [70]. In a recent work, Iwata *et al.* (2019) showed the retardance around the bubble using the birefringence for CTAB/NaSal solutions where they reported a velocity discontinuity at very small bubble volumes [71].

3. Materials and Their Rheological Characterization

3.1. Materials

3.1.1. Poly-acrylamide (PAAM)

The non-Newtonian solution is prepared with polyacrylamide (PAAM) AN SH and manufactured by SNF Floerger Turkey. The molecular formula of PAAM can be seen in Figure 3.1. Polyacrylamides are water-soluble polymers. They are widely used in drinking water production, wastewater treatment, mining enterprises, petroleum production, agriculture, textile, and cosmetic industries.



Figure 3.1. The molecular structure of PAAM.

Polyacrylamide is a high molecular weight polymer. The physical properties of PAAM can be seen from Table 3.1. Polyacrylamide is insoluble in any organic solvent except some acids (acetic, lactic), some hydroxy compounds (ethylene glycol, glycerol), some nitrogen compounds (formamide) and molten urea, and dissolves in a wide range of concentrations in water.

The viscosity of aqueous solutions increases with increasing molecular weight and decreases with increasing temperature. Viscosity in aqueous solutions of nonionic polyacrylamide is relatively unaffected by the electrolyte, and the hydrogen ion concentration [73,74]

Appearance	White powder		
Ionic character	Anionic		
Charge density	Medium		
Molecular weight (kg/kmol)	High (18E6-23E6)		
Dissolution time in Dl water @ 5 g/l @ 25°C (minutes)	60		
Storage temperature (°C)	0-35		
Shelf life (months)	24		

Table 3.1. The physical properties of polyacrylamide [72].

PAAM has a high solubility in water, however, it dissolves very slowly. While preparing the solution, PAAM should be added to the water, not vice versa. In cold water, dissolving PAAM is more difficult than in warm water. To dissolve PAAM in cold water, more time and agitation will be required.

During this research, it was found that the preparation of the PAAM solution is highly challenging. Even though PAAM is highly soluble in water, our trials showed that the usual mixing procedures do not work for the PAAM solution. Also, the information given in the literature about the preparation procedure is very limited. Below, one can find the procedure developed based on our observations and experience.

- (i) Distilled water is warmed until (32°C). If the temperature is too low, the dissolution rate is affected where the high temperature can cause deterioration in the polymer's structure.
- (ii) A mechanical mixer with blade marine impeller (Figure 3.2a) is chosen for proper shear. A different type of impeller with low shear can result in poor mixing, where an impeller with high shear can damage the polymer molecules.
- (iii) A vortex (Figure 3.2b) is created in the middle of the distilled water using the impeller at 1500 rpm speed.
- (iv) PAAM that is in powder form is slowly added in the middle of the created vortex. This step requires extra care as adding the polymer fast can cause the powder to



Figure 3.2. The blade marine impeller used for the preparation of the PAAM solutions (a) and the vortex created using the impeller (b).

swell and cohere to produce a mass. Once this mass is formed, the water will not be able to penetrate inside of it, and there will be lumps of polymer inside the solution.

- (v) When all of the powder is added into the vortex, the speed of the mix is reduced.
- (vi) The solution is mixed for only 60 minutes.
- (vii) The solution is transfer into a container with high surface area, and left there for 24 hours to remove the bubbles trapped inside the solution.
- (viii) The solution is stored at room temperature in a closed container, and kept away from the direct sunlight.
- (ix) Overall, it is possible to say that the rate of PAAM dissolution has a strong relationship with the amount used, the stirring speed, and the temperature.

3.1.2. Hydroxypropylcellulose (HPC)

Hydroxypropyl cellulose (HPC) is a derivation of cellulose that is highly soluble in water and organic compounds. Its molecular formula is given in Figure 3.3. HPC used in this work was purchased from Sigma Aldrich (CAS Number: 9004-64-2).

HPC is a liquid crystalline liquid that is in the cholesteric phase and highly bire-



Figure 3.3. The molecular structure of HPC.

fringent. In this work, HPC is added to the PAAM solution to make the solution birefringent. HPC solution is a shear-thinning fluid, showing viscoelastic behavior. Anisotropic solutions are strongly viscoelastic. The relationship between the birefringence and the flow shows that the flow birefringence increases with HPC concentration, with a significant increase as the liquid-crystalline phase is formed [75].

3.1.3. Glycerol

Glycerol is a highly viscous, hygroscopic substance that contains three hydroxyl groups in its structure, dissolving completely in water. It is a biochemical compound in terms of origin. The molecular formula of glycerol is $C_3H_8O_3$ and its IUPAC name is propane-1,2,3-triol where the molecular structure is given in Figure 3.4 [76]. In this study, glycerol is purchased from Isolab and is 85 % by weight. The calculation of the physical properties of 85 wt % of glycerol is based on the study of Cheng (2008) [77], and these values are tabulated in Table 3.2.



Figure 3.4. The molecular structure of glycerol.

Table 3.2. The physical properties of glycerol.

Appearance	Clear		
Form	Liquid		
Molecular weight (g/mol)	92.09		
Fraction of glycerol by mass	0.85		
Density (kg/m^3)	1218.4		
Dynamic viscostiy (Pa.s)	0.08 5		

3.1.4. Sodium dodecyl sulfate (SDS)

Sodium dodecyl sulfate (SDS) is an organic compound with surfactant properties. SDS, an organic salt, is classified in the group of organosulfate compounds. The molecular formula of SDS is $C_{12}H_{26}O_4SNa$ and its structure is given in Figure 3.5. It was purchased from Isolab and has CAS number of 151-21-3.

SDS is an anionic surfactant with a polar head sulfate group and apolar tail consisting of a 12 carbon alkyl chain. This tail gives the necessary amphiphilic properties.

Surfactants are adsorbed at the interfaces as a result of these structural units. Surfactant adsorption generally alters the surface or interfacial properties of the system. SDS molecule carries a negative charge in its active part; therefore it is called anionic.



Figure 3.5. The molecular structure of SDS.

3.2. Rheological Characterization

In this study, the rheological measurements were carried out using Anton-Paar MCR302 rotational rheometer at the Life Sciences laboratory of Bogazici University (Figure 3.6) and MCR92 at Anton Paar, Turkey. RheoCompass software of Anton Paar is employed during the measurements.



Figure 3.6. The MCR302 Anton Paar reometer at the Life Sciences Laboratory at Boğaziçi University.

Cone-plate probes (Measuring Cone CP25-2 and CP50-2) are used where the distance between the cone and the plate was 0.103 mm, and the diameter was 25 mm (CP25-2) and 50 mm (CP50-2). In Figure 3.7, the schematic representation of the cone and plate rheometer is demonstrated. In this figure, r_c is the cone radius

where θ and dc are the angle and the distance between cone and plate, respectively. In the measurements, the lower plate is fixed, and the upper cone can rotate at fixed or variable frequency values with a variable rotation number, fixed rotation number, or certain deflection angle amplitude for oscillating tests depending on the test type.

Probe name	Probe type	Angle (°)	Diameter (mm)
CP 25 - 2	Cone - plate	2	25
CP 50 - 2	Cone - plate	2	50

Table 3.3. The properties of the rheometer probes used for the rheological analysis.



Figure 3.7. Schematic representation of the cone-plate rheometer.

During the measurements, it is ensured that the sample temperature is kept constant everywhere by using a heating top cap. In addition, the excess solution overflowing from the measurement environment is removed in order not to cause measurement errors. Before starting the measurement, the normal force value affecting the measuring element is expected to be as close as to zero.

3.2.1. Rheological Characterization Tests

<u>3.2.1.1. Flow Curve test.</u> During this test, the change in viscosity depending on the shear rate or shear stress was observed. Flow curves are usually tested over a narrow

band of shear rate. Accordingly, solutions have been analyzed at 25°C over a shear rate range of 1-100 s⁻¹.

<u>3.2.1.2.</u> Amplitude Sweep Analysis. This test was performed to determine the viscoelastic properties of the solutions. The amplitude sweep analysis determined linear viscoelastic regions of the solutions, and the analyzes were carried out in this region. Graphs of G' (elastic) and G" (viscous) values against strain (%) are drawn.

<u>3.2.1.3.</u> Temperature Sweep Test. To be able to observe the effect of the temperature on the viscosity of the solutions, the temperature sweep test was used. For the samples, the temperature sweep test was performed at 0.01% strain within the linear viscoelastic region determined in the amplitude sweep test, at a constant frequency of 1 Hz and increasing with a speed of 5°C/min in the range of 20°C - 50°C.

3.2.2. Rheology Results of 85 wt% Glycerol

Glycerol is a Newtonian liquid and its rheology analysis for glycerol is given in the Figure 3.8. Although glycerol was purchased readily, no rheology analysis was available. Rheology analysis shows that shear stress and shear rate were directly proportional (Figure 3.8 - blue line). Although the viscosity of glycerol appears to increase with the shear rate, on careful inspection it can be seen that the change is in the order of 10E-3 (Figure 3.8 - orange line). Therefore, the viscosity of glycerol is determined to be constant with increasing shear rate, 0.085 Pa.s at 25°C as in [78].

3.2.3. Rheology Results of PAAM and PAAM+HPC Solutions

In this study, since the isothermal experiments were performed at 25°C, all of the rheological analyzes of the solutions were also performed at 25°C. Unless otherwise stated, all of results presented below are valid only for 25°C.



Figure 3.8. The flow curve of 85 wt% glycerol.

The rheological analysis of solutions containing only PAAM and PAAM+HPC are investigated. The concentrations used in the experiments that contained only PAAM were 0.10, 0.18, 0.25 and 0.40 wt%. The HPC concentration to be added in the birefringence experiments was tested with 3 different concentrations, i.e. 0.10, 0.20 and 0.40 wt% HPC. Birefringence was not observed enough in the trial experiments with 0.10 and 0.20 wt% HPC. For this reason, the HPC concentration was increased up to 0.40 w%. From our previous experiments, it was known that the birefringence was observed well with 0.5 wt% concentration of HPC. However, it was known that the higher the HPC concentration is, the higher the entire solution's viscosity is. As the overall viscosity increases, it becomes more difficult to perform the experiment due to the small bubbles trapped in the solution. Therefore, it is decided to keep the HPC concentration constant at 0.4 wt% while changing the PAAM concentration as 0.10, 0.18 and 0.25 wt%

As stated in the first test, the flow curve test, the relationship between viscosity of the solutions was examined depending on the shear rate; also the relation between shear stress and shear rate was studied. For all of the solutions it was found that shear stress increases with increasing shear rate (Figure 3.9 and Figure 3.10).



Figure 3.9. Shear stress, τ vs. shear rate, $\dot{\gamma}$ for different concentrations of PAAM solutions.



Figure 3.10. Shear stress, τ vs. shear rate, $\dot{\gamma}$ for different concentrations of PAAM + 0.40 wt% HPC solutions.

The solution with the highest concentration has the highest shear stress at a constant shear rate. Furthermore, the viscosity decreases with increasing shear rate (Figure 3.11 and Figure 3.12). At a constant shear rate, the solution with the highest concentration has the highest viscosity, i.e., 0.40 wt% PAAM in Figure 3.11 and 0.25 wt% PAAM + HPC in Figure 3.12.

The rheological analyses also shows that HPC had a significant effect on the viscosity. For example, the viscosity of 0.25 wt% PAAM solution is around 450 mPa.s at 1 s^{-1} shear where as it is around 2500 mPa.s for PAAM + HPC solution at the same shear rate. The flow curve test results show that all of the solutions are non-Newtonian and shear-thinning.



Figure 3.11. Viscosity, η vs. shear rate, $\dot{\gamma}$ for different concentrations of % PAAM solutions.



Figure 3.12. Viscosity, η vs. shear rate, $\dot{\gamma}$ for different concentrations of PAAM + 0.40 wt% HPC solutions.

Amplitude sweep test was applied to the solutions to obtain information about their viscoelasticity. During an amplitude sweep test, the amplitude of the shear stress is changed while the frequency is kept constant. For the analysis, the storage modulus G' and the loss modulus G" are plotted against the shear strain (Figures 3.13) and 3.14). The ratio of the two moduli (G' and G") gives information about the characteristic of the sample. First, the linear viscoelastic (LVE) region is found for all solutions. The LVE region shows the test region where the tests can be done without destroying the structure of the solution. All of the solutions show a constant plateau regime in the LVE region. When this region is investigated, it is seen that the storage modulus, G' is larger than the loss modulus, G". Therefore, it can be said that the solutions have gel-like structure. It is observed that there occurs a cross-over between storage and loss moduli. This transition shows where the solution starts behaving more like fluid than like gel. Again it is found that the addition of HPC into PAAM solution changes the structure of solution in terms of viscoelasticity.



Figure 3.13. Storage, G' and loss, G" modulus vs. shear strain, γ for different concentrations of PAAM solutions.



Figure 3.14. Storage, G' and loss, G" modulus vs. shear strain, γ for different concentrations of PAAM + 0.40 wt% HPC solutions.

The temperature-dependent variation of the apparent viscosity values was determined by applying the temperature sweep test. This test was carried out at a shear rate of 50 s⁻¹ as it is calculated as the average shear rate for our experiments. Temperature sweep test was carried out by providing a 5°C temperature increase per minute from 20°C to 50°C. The results show that the viscosity decreases as the temperature increases (Figure 3.15). It can be said that the average viscosity drop is around 25% for the solutions.



Figure 3.15. The effect of the temperature on the viscosity for different concentrations of PAAM solutions.

Finally, two different rheological models are applied, i.e. the power-law and the Carreau-Yasuda models, and tabulated in Table 3.4 and 3.5, respectively. Viscosity index, n, of the power-law equation given in Eq. 2.2 indicates how easily a fluid will deform under shear stress. In the Carreau-Yasuda model (Eq. 2.4), η_{∞} is the value taken by viscosity as the shear strain rate goes to infinity. λ is called relaxation time and is the inverse of the value of the shear strain at which the viscosity begins to decrease. η_0 is the zero viscosity value and is the value of viscosity when the shear strain rate is close to zero. λ and a are the relaxation time and Yasuda exponent, respectively. These rheology parameters are used in the calculation of the dimensionless numbers.

Solutions	n	K (Pa.s)	\mathbf{R}^2
0.10 wt% PAAM	0.73	0.03	0.99
0.18 wt% PAAM	0.46	0.31	0.99
0.25 wt% PAAM	0.44	0.51	0.99
0.40 wt% PAAM	0.34	1.45	0.99
0.10 wt% PAAM + 0.4 wt% HPC	0.57	0.46	0.99
0.18 wt% PAAM + 0.4 wt% HPC	0.48	0.93	0.99
0.25 wt% PAAM + 0.4 wt% HPC	0.41	1.97	0.99

Table 3.4. The rheological parameters for power-law model.

Table 3.5. The rheological parameters for Carreau-Yasuda model.

Solutions	η_0 (mPa.s)	η_{∞} (mPa.s)	λ (s)	n	a	\mathbf{R}^2
0.10 wt% PAAM	14	2.0	0.1	0.52	1.1	0.99
0.18 wt% PAAM	679	3.3	2.4	0.36	1.2	0.99
0.25 wt% PAAM	834	3.7	3.0	0.39	1.1	0.99
$0.40 \ \mathrm{wt}\%$ PAAM	7105	5.0	8.4	0.29	1.5	0.99
0.10 wt% PAAM + 0.4 wt% HPC	2552	0.1	34.3	0.55	0.5	0.99
0.18 wt% PAAM + 0.4 wt% HPC	6595	4.4	36.8	0.46	2.8	0.99
0.25 wt% PAAM + 0.4 wt% HPC	18829	0.1	45.2	0.41	4.9	0.99

4. Experimental Setups and Methods

The liquid column is made of glass. The glass plates (0.50 cm thickness) are glued together with the aquarium silicone (Sista). Later, a transparent liquid rubber (Leak Fix Spray Rubber Transparent) is applied to the glued areas. At least four layers of spray liquid rubber application are required. The places that will not be covered are carefully masked with paper tape. After one hour of application, the liquid rubber is hardened, and the liquid column becomes leakproof. The liquid rubber can withstand temperatures between -40 ° C and +90 ° C. This temperature range is much wider than the temperatures employed in non-isothermal experiments. The liquid column dimensions are 12 cm x 12 cm x 50 cm (Figure 4.1a).



Figure 4.1. The view of liquid column (a) and the plumbing system (b).

Under the liquid column, there are a plumbing system (Kale Armatür) and hose (Evci Plastik) (Figure 4.1b) and a syringe with a volume of 60 ml (SET Plastic Syringe) connected to this hose. At the beginning of the experiment, the syringe is filled with air. During the experiment, a bubble is obtained by manually pushing air from the syringe. Although this method is not fully controlled, bubble size with a wide range of volumes can be obtained. A syringe, directly connected to the liquid column is used

in another liquid column to obtain bubbles with smaller sizes. With the needle (21G) coming out of this syringe, bubbles are sent into the liquid directly.

4.1. Experimental Setup for the Birefringence Experiments

This section deals with the optical setup to analyze the bubble dynamics, i.e., the shape, the speed and also the birefringence experimental setup.

4.1.1. Optical Experimental Setup

The experimental setup is shown in Figure 4.2. In order to ensure a stable and easy optical alignment, two interlocking rails (Thorlabs – XT66SD-750) are mounted on the optical table (B6090AX), and the optical materials are mounted on these rails. In order to align the laser (CNI Lasers – MLL-III-637-100mW) light on the rail, kinematic frames numbered 4 and 5, with mirrors inside (Thorlabs – PF10-03-P01 inside KM100), are used as "Z-Fold" configuration as shown in Figure 4.3.

First of all, the target (Thorlabs – LMR1AP) is placed on the rail with the help of the optical post (Thorlabs – TR300-M), the post holder (Thorlabs – PH150-M), the rail carrier (Thorlabs – XT66P2-M) and the lens mount (LMR1) at the height level where the birefringence around the bubbles will be measured. After fixing it at 35 cm height from the optical table, the target is slid on the rail and brought to the closest position to the laser. Here, by manipulating the two knobs of the kinematic frame numbered 4, it is ensured that the laser coincided with the target. Later, the target is shifted to the farthest position from the laser on the rail, and this time, the laser is brought to the target with the help of the knobs of the kinematic frame numbered 5. The target is brought close to the laser again, and this process continues until the laser does not deviate from the target when the target is at either end of the rail. Thus, the alignment of the laser beam on the optical table and the rails is achieved.



Figure 4.2. Optical setup. (1) Optical table, (2) Optical rail system, (3) Laser, (4-5)
Kinematic frames and mirrors, (6) Plano-concave lens, (7) Plano-convex lens, (9)
Liquid column, (8-10) Polarizers, (11) Camera, (12) Illuminator.



Figure 4.3. The Z-Fold configuration for the alignment of the laser light.

Two optical lenses and their frames (Thorlabs – LMR2 -M and LMR6-M) are used to bring the laser beam to the appropriate size. One of these lenses is plano-concave (Thorlabs – LC1975-A), and the other one is plano-convex (Thorlaabs – LA1380-A). In order to check the alignment of the optical lenses with the laser beam that is previously aligned with the rail system, the lenses are placed on the rail starting from the farthest end of the laser. Therefore, first of all, the plano-convex lens, number 7, is placed on the rail with the flat side of the lens facing the laser, and it is checked whether the reflected laser light was symmetrical, smooth, and in line with the incoming laser light. If these conditions are not met, the alignment of the lens is completed by changing the angle and the height of the lens. The same procedure is applied to the plano-concave, number 6 lens that is positioned closer to the laser on the rail. However, the flat side of this lens is positioned so that it faces the plano-convex lens, number 7, instead of facing the direction of the laser beam. In this way, the laser beam is aligned according to the column criteria and in a collimated manner.

Finally, polarizers (polarizer and analyzer with 2 in diameter, Thorlabs – LPVISE 200-A) are installed in high precision rotating frames (Thorlabs – PRM2-M) with polarization directions perpendicular to each other. The liquid column is placed on the rail system in between the polarizers after the lenses as shown in Figure 4.2. In order to perform birefringence analysis, the camera (Nikon D7200) is placed at the end of the optical assembly using a tripod stabilizer, after the polarizer number 10.

4.1.2. Adjusting the Light Distribution

The transverse mode of the laser used in the experiments is Near TEM_{00} , and its M2 value is 1.135. Although lasers with this mode are more affordable than the lasers in the TEM_{00} mode, the shapes of the beam spots are more distorted and far from Gaussian. Their shape is elliptical rather than circular since one axis has more openings than the other. This one-way opening causes the shape of the beam spot in that direction and the edges of the lenses inside the laser to be cropped due to the diffraction effect. The sample image of a laser from the literature [79] and the image



Figure 4.4. NearTEM₀₀ Beam from the literature [79] and this study.

of the laser used in the experiments with TEM_{00} mode are given in Figure 4.4.

This clipping in the shape of the laser beam spot is a factor that adversely affects the birefringence experiments. Magnifying the laser beam and using the mediumintensive part of the beam to get rid of this clipping is one of the methods. This problem has been solved in this study by magnifying the laser beam since magnification is also necessary for birefringence experiments. A small number of defects in the middle region were also eliminated by the laser beam being homogenized as it passes through the solution and matching the location of bubbles with the middle part of the beam for the birefringence analysis.

4.1.3. The Procedure for the Birefringence Experiments

The Nikon D7200 camera is used in high resolution (1920x1080) to record the birefringence experiments. Videos are captured at 60 fps. Since the polarizer is perpendicular to the analyzer at the beginning of the experiment and no bubble passes through the liquid at that time, no optical axis difference occurs in the liquid. Therefore, the laser beam passing through the polarizer cannot pass through the analyzer (Figure 4.2) since it is parallel to the polarizer and perpendicular to the analyzer; thus, a dark image like in Figure 4.5a is obtained at the beginning of the experiment. When the bubble rises in the column and reaches the measurement area, a difference

in the optical axis occurs due to the angle change between the liquid molecules. In this way, the laser beam passing through the polarizer changes its direction while passing through the liquid, and since every component of the laser beam is no longer perpendicular to the analyzer, it succeeds in passing through the analyzer. As a result, a light formation is observed around the bubble (Figure 4.5b).



Figure 4.5. The dark image (a) obtained at the beginning of the birefringence experiment and the birefringence image as the bubble passes(b).

As it is mentioned before, the polarizer and the analyzer are perpendicular to each other throughout the entire experiment. The birefringence experiments are performed twice. However, in the second experiment, both the polarizer and the analyzer were shifted by an angle of 45°.

The image series obtained from a birefringence experiment can be seen in Figure 4.6. The experiment shown in Figure 4.6 was overexposed for the visual quality. Due to the overexposure, a completely dark image has not been obtained in this series of experimental images as mentioned for Figure 4.5a. A relatively darker view is obtained in Figure 4.6a. Then, in Figure 4.6b-p, the laser beam that has managed to pass through the polarizer can be seen as a result of the bubble motion.

4.2. Experimental Setup for the Bubble Dynamics

A high-speed camera (Optronis CamRecord CR600x2) is used in the experiments for volume-velocity analysis of the bubbles. The camera resolution is 1280 x 1024 pixels and can record a maximum of 500 fps. This camera is positioned perpendicular to the laser beam, facing the illuminator and the liquid column. The drawing of the idiomatic mechanism of bubble dynamics experiments is given in Figure 4.7. For simplicity, the optical part is not shown in detail. Single color LED continuous light (Mcoplus LE520A) is preferred for the illumination.

The light intensity of the illuminator and homogeneous the reflection of this light onto the liquid in the column are essential in terms of the boundaries of the bubbles along the column. For this reason, the light skate is covered with a diffuser. Because the light is perpendicular to the camera, the bubble boundaries become more pronounced. Bubbles with clear boundaries could be separated and analyzed easily by the volumevelocity analysis algorithm.

a)	b)	c)	d)
e)	í)	g)	h)
i)	D C	k)	D C
m)	n)	o)	p)

Figure 4.6. The image series obtained from a birefringence experiment.

The camera is controlled from a computer using TimeBench 2.6.30 software. The camera is operated via a trigger. In the experiments, image ROI trigger sensitivity setting is employed where the camera starts recording as the brightness of the specified area changes. Therefore, an area in the frame where the bubble will pass is chosen, and the camera triggers when the brightness changes in this area by 90%. Thanks to the multi-segment feature of the camera, a separate video is shot for each bubble, and finally, they are turned into a single video by the software. Thus, high-speed processes such as bubble movement can be recorded, and less memory is used by not recording the moments when the bubble does not pass.



Figure 4.7. The sketch of the bubble dynamics experimental setup. (3) laser, (9) liquid column, (11) birefringence camera, (12) light source, (13) high-speed camera.

4.3. Experimental Setup for the Schlieren Method

The Schlieren imaging technique captures density gradients that are usually invisible in a transparent medium such as air, water, and glass. While Schlieren imaging is often used for laboratory-based in-depth studies, the technique known as shadowgraphy is more commonly used in field studies. When a light ray hits an environment, variations in the density gradient change the direction of the light to make it visible. There are different types of Schlieren method, such as v-, y- or z- type systems, based on the system's layout. In this study, the v-type system is used, and the system drawing is shown in Figure 4.8.



Figure 4.8. The scheme of the v-type Schlieren method.

The v-type Schlieren device employs a point light source to generate a beam of light. However, instead of the optical lenses used in other types, the v-type device employs a concave mirror to focus the light back to a single point. At that point, the razor edge cuts off the light that comes directly while allowing the refracted light to pass around the razor captured by an imaging device [80]. The light source and the razor edge are placed at twice the focal length of the concave mirror, resulting in the incoming light being reflected to the razor.

The image obtained from a demonstration of the Schlieren experiment is given in Figure 4.9. In this figure, there is a single plume of a lighter. In v-type Schlieren setups, the double imaging situation can cause problems if the focal length of the concave



Figure 4.9. The double view of Schlieren image of the single lighter plume.

mirror is short, which causes the beam light to be refracted twice. A beamsplitter cube is used to remove the double image formation. Even tough, this modicification worked, it caused other problems. Therefore, the double view problem is kept and it was only overcome by adjusting the distance of the camera from the mirror, increasing the double images from each other and cutting the view into two during the video analysis.

4.4. Experimental Setup for Non-Isothermal Experiments

For the nonisothermal experiments, a different liquid column was designed. This column's one wall is made of aluminum and attached to a water bath where the temperature is always kept constant at 25°C. A heating plate is placed next to the opposite wall. The scheme of the designed liquid column can be seen in Figure 4.10. The temperature is monitored at different spots (shown as red dots in Figure 4.10) using thermocouples. The bubble dynamics are investigated as explained in Section 4.2.


Figure 4.10. The scheme of the liquid column designed for the non-isothermal experiments.

4.5. Surface Tension Measurement

Du Nuoy ring, a very common method, is used to measure the surface tension of the solutions with and without surfactants. A platinum-iridium ring is submerged in a liquid and then the liquid is drained from the beaker. The force required for the ring to pull up the meniscus of the liquid is proportional to the surface tension [81–83]. The experimental setup used for the Du Nouy method is given in Figure 4.11.

First, the ring is immersed into the solution (Figure 4.12-2), where the torsion dynamometer is set to 0. Then, the solution is drained from the bottom of the beaker. As the solution is drained, the lever arm of the torsion dynamometer is readjusted to balance the white line that is shown on the dynamometer. The value read on the torsion dynamometer is recorded when the Du Nouy ring is separated from the solution's surface (Figure 4.12-4). This value is the force, F that acts along the edge of the length, l, which is parallel to the surface to keep the film [84,85].



Figure 4.11. The sketch of the Du Nouy ring method experimental setup [85].



Figure 4.12. The process of Du Nouy ring method.

The surface tension, γ can be calculated as

$$\gamma = \frac{F}{\ell},\tag{4.1}$$

where ℓ is the length of edge and given as

$$\ell = 2 * 2 * \pi * r_r. \tag{4.2}$$

Here, the diameter of the ring, r_r used in the experiment is 19.65 mm.

It turns out that the surface tension of the 0.25 wt% PAAM solution is 74 mN/m, which is close to the value of water, 72 mN/m at 24°C [86]. The results also show that the surface tension decreases with increasing surfactant, SDS concentration. The surface tension of the solutions are given in Table 4.1.

 Solution Type
 Surface Tension (mN/m)

 0.25 wt% PAAM
 74.0

 0.25 wt% PAAM + 10 ppm SDS
 66.8

 0.25 wt% PAAM % 100 ppm SDS
 45.2

Table 4.1. The results of the Du Nouy Ring's method at 25 °C.

5. EXPERIMENTALLY MEASURED PARAMETERS

In this section, the measured parameters are explained in detail before presenting the results. The section includes two sub-sections for bubble dynamics and birefringence measurements.

5.1. Measured Parameters for the Bubble Dynamics

Here, first the bubble diameter and volume calculations are shown. Then, the bubble shape parameters are explained. Finally, the dimensionless numbers required to study and understand the bubble dynamics are presented.

5.1.1. Bubble Equivalent Diameter, Volume and Velocity

5.1.1.1. Bubble Equivalent Diameter, d. In the equivalent diameter method, the recorded image of the bubble is analyzed with the edge detection algorithm that will be mentioned in Section 6.2.1. The volume (V) of the bubble is obtained, then a diameter calculation is made for this area.

Equivalent diameter (d) is the diameter value of this bubble if it was spherical and calculated as follows,

$$d = \sqrt[3]{\frac{6V}{\pi}}.$$
(5.1)

5.1.1.2. Bubble Volume, V. At first, the rotating solid method was applied to calculate the bubble volume. However, this method was time consuming and not sensitive as it was required. Therefore, the truncated cone method is adopted. The shape of the bubble is considered to be axisymmetric with respect to the major axis of the bubble profile. The bubble is divided into multiple groups of small truncated cones as shown in Figure 5.1. The volume of each (ith) cone is calculated by the formula given below,

$$V_i = \frac{\Pi h}{3} \left(R^2 + r^2 + Rr \right) \tag{5.2}$$

where R and r are the radius of the lower and upper sections, respectively; and h is the height of the cone. These three variables can be derived directly from bubble profiles [87]. Finally, the total volume of the bubble is the sum of all the smaller cones (Figure 5.1).



Figure 5.1. The bubble profile used to determine the volume with the truncated cone method (reprinted with permission from [37]).

5.1.1.3. Bubble Velocity, $u_{.}$ The change in the position of the bubble is used to calculate the bubble velocity, u_{b} . First, the coordinates of the center of mass are determined

for each video frame, and the difference in the center of mass coordinates, x_i between the two consecutive frames of the bubble, is recorded and divided by the time difference between the frames, Δt . Thus, the average bubble velocity can be obtained through the liquid column as

$$u_b = \frac{x_i - x_{i-1}}{\Delta t}.\tag{5.3}$$

The terminal bubble velocity, u_B is calculated as the average value of the velocity values, u_b above the distance where the bubble velocity seemed to reach a steady value. It was made sure that the change in the bubble velocity is less than 5% and at least 26 frames are used to calculate the average terminal bubble velocity, u_B .

5.1.2. Bubble Shape Parameters

5.1.2.1. The Ratio of a/b. It is the ratio of the minor axis, a to the distance, b from the center of gravity to the tip of the bubble's tail (Figure 5.2a). The principal axis is the end points of the longest line that can be drawn through the bubble. The major axis length of a bubble is the pixel distance between the major axis endpoints, which is the distance between the bubble's apex and the tip of the bubble's tail. The minor axis is the end points of the longest line that can be drawn on the bubble while remaining perpendicular to the main axis. The minor axis length of an object is the pixel distance between minor axis endpoints.

<u>5.1.2.2. Extent.</u> It is the ratio of the area of the bubble (S5) to the area of the bounding box of the bubble (S1+S2+S3+S4+S5) (Figure 5.2b). The bounding box of a bubble is a rectangle surrounding the bubble. The dimensions of the bounding box are the dimensions of the major and minor axes.

$$Extent = \frac{S5}{S1 + S2 + S3 + S4 + S5}$$
(5.4)



Figure 5.2. The bubble shape parameter: the a/b ratio a), extent b) and solidity c).

5.1.2.3. Solidity. This parameter defines whether a shape is convex or concave (Figure 5.2c). It can be found by dividing the area inside the bubble into the area surrounded by a convex body (convex hull). The convex body (convex hull) can be thought of as a rubber band stretched around the bubble seen as red line in Figure 5.2c. The solidity of a completely convex shape is 1. The more the solidity deviates from 1, the greater the degree of concavity in the structure is (Figure 5.2c).

<u>5.1.2.4. Circularity.</u> It mathematically shows the degree of resemblance to a perfect circle. A value of 1 means a perfect circle. As the roundness value deviates from 1, the shape becomes less circular.

The circularity parameter is calculated as

$$Circularity = 4\pi \frac{Bubble area}{Bubble perimeter^2}.$$
 (5.5)

<u>5.1.2.5. Eccentricity - 1, E1.</u> It is the ratio of the length of a bubble's short (minor) axis to the length of its long (major) axis.

5.1.2.6. Eccentricity - 2, E2. It is defined as the ratio of equivalent diameter to the short (minor) axis.

5.1.3. Dimensionless Numbers

Dimensional numbers show how the system behaves and allow comparisons between different systems. The dimensionless numbers used in this study are as follows:

(i) Reynolds number, Re: It is the ratio of the inertial forces to the viscous forces.
Chhabra (1988) [7] and Lali *et al.* (1989) [88] defined the average shear rate over an entire particle as

$$\dot{\gamma} = \frac{u_B}{d} \tag{5.6}$$

where $\dot{\gamma}$ is the shear rate, u_B the terminal velocity, and d is the equivalent diameter given in Eq. 5.1.

The apparent viscosity, η for power-law fluids is defined as

$$\eta = K \left(\frac{u_B}{d}\right)^{n-1}.$$
(5.7)

Here, n is the flow behaviour index (dimensionless) and K is flow consistency index (Pa.sⁿ). Finally, Re for a power-law fluid becomes,

$$Re = \frac{\rho u_B^{(2-n)} d^n}{K}.$$
(5.8)

(ii) Bond number, Bo: It is a dimensionless group used for the analysis of the behavior of bubbles and drops [3]. In the literature, it is sometimes referred to as the

$$Bo = \frac{\rho d^2 g}{\sigma} \tag{5.9}$$

where σ is the surface tension of the liquid and g the gravitational acceleration [3].

(iii) Capillary number, Ca: It is a number that relates the viscous forces in the system to the surface tension forces and is given as [3]

$$Ca = \frac{\eta u_B}{\sigma}.\tag{5.10}$$

(iv) Weber number, We: It is the ratio of inertial forces to surface tension forces and given as

$$We = \frac{\rho du_B^2}{\sigma}.$$
(5.11)

(v) Drag coefficient, C_D : It is used to measure the friction or resistance of a bubble or droplet rising in a liquid medium. There are several ways to calculate C_D . It can be calculated experimentally for a rising bubble as

$$C_D = \frac{4}{3} \frac{dg}{u_B^2} \left(\frac{\rho_\ell - \rho_g}{\rho_\ell}\right). \tag{5.12}$$

According to Hadamard-Rybczynski' law [89, 90] for Re < 1 [18], the drag coefficient for deformable bubbles is

$$C_{D,2} = \frac{16}{Re}.$$
 (5.13)

Using Hadamard-Rybczynski' law, Margaritis *et al.* (1999) empirically obtained C_D for shear-thinning fluids for Re < 60 as [32,91]

$$C_{D,3} = \frac{16}{Re} \left(1 + 0.173 R e^{0.657} \right) + \frac{0.413}{1 + 16300 R e^{-1.09}}.$$
 (5.14)

5.2. Measured Parameters for the Birefringence

The polarization of the light is given in Figure 5.3. The stress-optical law is used in the retardance, δ (phase delay) and the stretching angle, χ calculations. The equations used for the analyzes are given below. The calculated retardance, δ will be a depth average value for the bubble volume.



Figure 5.3. The polarization of light as it passes through a birefringent medium.

The intensity of light passing and transmitted through a birefringent medium is denoted by I (θ) and is shown as follows, using the stress-optic law as

$$I(\theta) = I_0 \sin^2([\chi + \delta]) \left(\frac{\delta}{2}\right).$$
(5.15)

Here I_0 indicates the intensity of the incident light beam and its calculation is shown in Section 6.1. I_0 is constant throughout the experiment for a given solution concentration. I_0 needs to be studied and adjusted for each concentration value.

The experiments are repeated at two different angles. In the first case, the polarizer and the analyzer are perpendicular to each other. In the second case, the polarizer and the analyzer are again perpendicular to each other; however both are rotated 45° in the same direction compared to the first case.

When both of the experiments are performed, the bubbles with the same volume from each experiment are matched with each other where the intensity of the first case is called $I(\theta)$ and the second one is called $I(\theta + 45)$.

The retardance, δ can be calculated as

$$I(\theta) + I(\theta + 45) = I_0 \sin^2\left(\frac{\delta}{2}\right).$$
(5.16)

If $\theta = 0$ in Eq. 5.15, the equation becomes

$$\frac{I(\theta)}{I(\theta+45)} = \tan^2(2\chi),\tag{5.17}$$

and χ can be calculated from here.

The retardance, δ is directly proportional to the birefringence, Δn as it is given in Eq. 5.18 where h and λ_i indicate the optical path and the wavelength of the incident light, respectively.

In this study, the optical path is the distance that light travels through the polymer solution; i.e. the column width of 12 cm. λ_i indicates the wavelength of the incident light, and its value for the laser used in the experimental setup is 637 nm.

The birefringence can be calculated from the following correlation as

$$\delta = \frac{2\pi h}{\lambda_i} \Delta n. \tag{5.18}$$

6. IMAGE PROCESSING AND ANALYZING TECHNIQUES

6.1. Intensity Calibration

When the birefringence experiments were performed, it has been noticed that the pixel values obtained from the birefringence photographs recorded with the camera did not reflect the actual light intensity coming to the camera sensor; and it would be wrong to use these pixel values directly in the optical stress law. Therefore, there is a need for the calibration of the intensity.

The reason for the mismatch between the intensity and the pixel value is that while converting the lossless raw data format, recorded by the camera, to non-lossless formats such as the *jpeg* extension, the camera uses RGB values and calculates according to a formula predetermined by the company for the camera, i.e., Nikon in this study [92]. For this reason, the actual light intensity - average pixel value calibration was performed separately for the exposure times of the camera that are used in the experiments.

For this purpose, the intensity of the laser light coming to the camera was measured with a light intensity sensor (Thorlabs/PDA55) that is placed right in front of the camera, and then the laser light was photographed with the camera. Then, using ND filters (Thorlabs/NDC-50C-4M) which are known to reduce the light intensity with a specified ratio, an average of about twenty-five photographs of the laser light, which can vary in number according to the exposure setting, were taken. As an example, sample photos with decreasing light intensity taken at 1/200 sec exposure time are shown in Figure 6.1. The photographs were processed in MATLAB, and the average pixel value was examined by selecting the same area in each frame where the laser light is visible and smooth. Thus, both the actual light intensity coming into the camera in each photo and the average pixel value of the image corresponding to that light



Figure 6.1. Sample calibration photos for the 1/200 sec exposure time where each frame represents the photo of the laser light taken with increasing filter.

intensity was obtained. A graph similar to that in Figure 6.2 is plotted for each of the exposure times measured using the collected data.

As seen in the literature, the relationship between the intensity versus pixel value is not linear, but a cubic polynomial [93]. For example, the response curve of selected digital cameras are shown in the literature [94]. For calibration, it is sufficient to obtain the response curve for each exposure time to be used in the birefringence experiments. After the experiment, in order to obtain the actual light intensity, this calibration curve is used. Since the density of the liquid in the column and the applied laser power may vary from experiment to experiment due to the concentration of the solution, the I₀ light intensity in the formulas will also change. In order to use the correct I₀ value, only the polarizer and the analyzer are removed before each experiment, and the image of the camera and the laser light are recorded. Then, the actual value of I₀ is calculated using the response curve belonging to the exposure time at which this image is recorded.



Figure 6.2. The calibration graph for the 1/200 sec exposure time.

A critical issue to be considered in terms of photographing the light intensity in the experiments is overexposure. In cases of overexposure, birefringent measurements will be inaccurate because the pixel values reach a maximum value of 255 (for 8-bit) when the camera exposure time or the laser power is high. Since the pixel values should decrease or increase according to the changing parameters of the bubble or the solution, the pixel values will not undergo any change in case of overexposure, and the analysis using these pixels will also be wrong. The presence of the overexposure is checked with the histogram feature in Image J of the photos recorded using large bubbles at the beginning of each experiment. The overexposure problem is solved by adjusting the power of the laser in case of overexposure. A sample photograph with overexposure (Figure 6.4a) and the histogram measurement of the chosen region (circled with blue on Figure 6.4b) where the birefringence is observed in this photograph are given. As can be seen from the histogram graph, there are about 1494 overexposed pixels.



Figure 6.3. Sample photo with overexposure (a) and the histogram analysis of the circled area (b).



Figure 6.4. Sample photo when the overexposure is removed (a) and the histogram analysis of the circled area.

6.2. Image Analysis

6.2.1. Image Analysis for Bubble Dynamics

MATLAB library functions are used for the edge detection algorithm of the bubbles. The algorithm used for the edge detection and to measure bubble properties, i.e. volume, velocity and shape parameters, are as follows:

Calibration:

- Entering the fps (frame per second) information of the experiment video after running the code and using *imdistline* on the ruler image (taken for the calibration purposes at the beginning of each experiment by placing a ruler in the liquid column) in the first frame.
- Choosing the region of the bubble's movement path by cutting it with *imcrop*.
- Determining the position of the label (a label with a specific size is placed on the edge of the liquid column) to be able to remove possible vibrations by cutting with *imcrop* later in the algorithm.

Bubble creation:

- Running the *imbinarize* code whose *adaptive threshold* feature is selected to detect bubbles in each frame.
- Using the *imfill* function to fill in the middle of bubbles that is caused by the light shining on the bubble surface.
- Binary recovery of all bubbles in each frame.

Filtering:

- Using *bwareafilt* to get the bubble with the largest area in a frame.
- Using *bwareaopen* to filter the bubbles with an area below than a specified value to eliminate very small bubbles.

Calculation of bubble parameters:

- Obtaining bubble area with *bwarea* and calculation of the equivalent diameter.
- Determining the coordinates of the center of mass of the bubble with *regionprops*, numbering the bubble and finding other geometric properties.

- Removal of position and velocity shifts caused by possible vibrations from the parameters that is found using the label mentioned above.
- Finding bubble perimeter with *bwboundaries*.
- Finding the volume using the truncated cone volume method.
- Finding the shape parameter, β by adapting the parametric equation to the environment.
- Finding the convex body around the circumference with the *convhull*.
- Finding the major and minor axes and the angles of these axes with the *bwferet* function.

Finalizing:

- Matrix and vectorial recordings of all analysis results.
- Visual recording of the results with *writeVideo*.

6.2.2. Image Analysis for Birefringence Measurements

In order to perform birefringence analysis, the photograph of each bubble must first go through some processes. The captured video is divided into frames by a software (Free Video to JPG), and each frame of each bubble observed during the experiment is extracted as in Figure 6.5a. Besides, the initial frame of the video is extracted to be used as the background frame (Figure 6.5b). The background image is subtracted from the bubble image (Figure 6.5c). Finally, the image (Fig 6.5c) is converted to grayscale, i.e., 8 bits. ImageJ program was used for all of these processes.



Figure 6.5. The image of a bubble obtained in the birefringence experiment (a), the background image (b), the image of the bubble where the background is removed(c) and the 8-bit grayscale version of this image (d).

The bubble images are analyzed using MATLAB. As mentioned in Section 5.2, the experiments are performed for a second time by rotating both the polarizer and the analyzer in the same direction for 45°. From each experiment, two intensity values are obtained. When the bubbles with the same volume are matched from both experiments i.e., the first intensity value is named as $I(\theta)$ and the second intensity where the polarizer and analyzer are shifted by 45° is called $I(\theta + 45)$. As bubbles are assumed to be symmetrical, the measurements are performed only for the left side of the bubble. The retardance, δ and the polymer stretching angle, χ are calculated using the equations given in Section 5.2, $I(\theta)$, $I(\theta + 45)$ and I_0 (explained in Section 6.1).

7. RESULTS AND DISCUSSION

7.1. Isothermal Experiments

7.1.1. Control Experiments with Glycerol

Even tough, this work is only interested in non-Newtonian solutions, the experiments are also performed with a Newtonian solution, i.e. glycerol. To test the experimental setup by comparing the results with the literature. The average terminal velocity of the bubbles are measured and drawn against the bubble equivalent diameter to study the dynamics of an air bubble rising in glycerol; and shown in Figure 7.1.



Figure 7.1. The average terminal velocity of a bubble rising in 85 wt% glycerol as a function of bubble equivalent diameter.

Iwata *et. al* (2019) [71] stated that the average terminal velocity and the bubble diameter are quadratically related for Re < 1 which is the Stokes region. The Re for the data in Figure 7.1 is between 2 and 40. It was found that the average terminal velocity is quadratically proportional to the equivalent diameter for this study as well. To compare our results with the literature [95, 96], first the viscosity of glycerol is measured and reported in Section 3.2.2. The comparison is given in Figure 7.2. The results are close to each other and matching in terms of behaviour. Specifically, it can be seen that the results are closer to each other with increasing bubble equivalent diameter.



Figure 7.2. The average terminal velocity of a bubble rising in 85 vol% glycerol solution as a function of bubble equivalent diameter. Comparison with the literature [95, 96].

7.1.2. Bubble Dynamics for Different PAAM concentrations

To study the effect of the concentration on the bubble dynamics, four different concentrations, i.e., 0.10, 0.18, 0.25 and 0.40 wt%, are employed. At this point, it is necessary to add that the legends of 0.10 wt% PAAM, 0.18 wt% PAAM, 0.25 wt% PAAM and 0.40 wt% PAAM are labeled as 0.10 PAAM, 0.18 PAAM, 0.25 PAAM, 0.40 PAAM, respectively to prevent the crowdness in the graphs.

For 0.10 wt% PAAM solution, bubbles larger than 100 mm³ showed a 3D motion, which is captured as the volume is investigated as a function of time, i.e., the column length (Figure 7.3). Bubbles larger than 100 mm³ showed wobbling and hence these

bubbles are not considered in the steady-state results. Consequently, for the 0.10 wt% PAAM solution, the terminal velocity results (Figure 7.4) include bubble volumes up to 100 mm^3 .



Figure 7.3. The bubble volume as a function of time for 0.10 wt% PAAM solution.

In Figure 7.4, the average bubble terminal velocity is drawn against the bubble volume for 0.10, 0.18, 0.25 and 0.40 wt% concentrations. It is observed that for a given polymer concentration, the bubble terminal velocity increases as the bubble volume increases. Until 300 mm³, the polymer concentration has an effect on the average terminal velocity. The results show that, the average terminal velocity of a bubble increases and makes a plateau around 250 mm/s for all concentrations. Bubble terminal velocity reaching a plateau is an expected behaviour regardless of the polymer type or concentration and matches with the literature [30].

It is found that the terminal velocities for 0.18 and 0.25 wt% PAAM are very close to each other and they even coincide with each other for bubble volumes larger than 20 mm³. This result is expected as the viscosity of both solutions are very close to each other for the shear rate of interest (50⁻¹ in order of magnitude, Figure 3.11), including the zero shear rate viscosity, η_0 . The results also show that 0.10 wt% solution has the highest average bubble terminal velocity for the limited bubble volume range.



Figure 7.4. The average terminal velocity as a function of the bubble volume for different concentrations of PAAM solution.

Finally, no evidence is found regarding the discontinuity in the bubble terminal velocity. There is no concensus in the literature on the presence of discontinuity for the bubble terminal velocity at a critical volume (Table 7.1). The results clearly indicate that experiments performed with PAAM of certain brands, e.g. Floerger (also used in this dissertation), Sigma-Aldrich and Sinopharm, show no discontinuity. However, a discontinuity in the velocity is detected for Separan AP-30, Praestol and P2540 brands of PAAM. Therefore, one might say that the discontinuity is directly related with the manufacturing of the polymer.

Besides the velocity, we are also interested in bubble shapes. The parameters and their definitions are given in Section 5.1.2. The images, the bubble dynamics parameters and the dimensionless numbers of the bubbles are tabulated in Table 7.2 for 0.25 wt% PAAM solution and Table 7.3 for 0.40 wt% PAAM solution. In these tables, V, d, u_B , a, b, C, Ext, Sol, E-1, E-1, E-2 are volume (mm³), equivalent diameter (mm), the minor axis (mm), the distance from center of gravity to the tail (mm), circularity, extent, solidity, eccentricity 1 and 2, respectively.

Study	PAAM polymer concentration, type and brand	Discontinuity
[18]	0.25, 0.50 and 0.75 wt % AN 913 SH -Floerger	No
[20]	0.25, 0.50 and 0.75 wt % AN 913 SH - Floerger	No
[16]	0.5 and 1.0 wt % AN 905 SH - Floerger	No
[35]	0.6, 0.8 and 1.0 wt% AR - Sinopharm	No
[37]	0.10, 0.25, 0.50 1.00 and 1.25 wt % Sigma Aldrich	No
[15]	0.15, 0.20 and 0.25 wt % Separan-AP30	Yes
[14]	0.5, 1.0 and 1.5 wt% - Separan AP3	Yes
[97]	0.5, 1.0 and 1.5 wt% - Separan AP30	Yes
[58]	0.075,0.1,0.15,0.2 and 0.25 wt $%$ - Separan AP-273	Yes
[40]	0.05, 0.10, 0.15, 0.20 and 0.25 wt % - Separan AP-273	Yes
[40]	0.5 and 1 wt % Separan AP- 230	Yes
[44]	0.1, 0.3, 0.8, 1 and 0.05 wt % Praestol 2500	Yes
[44]	0.1, 0.3, 0.8, 1 and 0.05 wt % - P2450	Yes

Table 7.1. The discontinuity in the bubble terminal velocity: literature that used PAAM.

The first shape parameter investigated here is the extent of the bubble. The extent is defined as the ratio of the bubble's area to the area of the bubble's bounding box as defined in Eq. 5.4 and shown in Figure 5.2b. This parameter is used to obtain information about how the area of a bubble changes and to observe if the concavity of a bubble can be tracked. The results show that the extent decreases with increasing concentration for a given bubble volume (see the bubble volume with $\sim 300 \text{ mm}^3$, extent decreases from 0.74 to 0.70 in Tables 7.2 and 7.3, respectively).

The results show that the extent decreases until $\sim 25 \text{ mm}^3$ which means that the bubble's area decreases compared to its bounding box. This can be explained by the bubbles turning from spherical to the prolate shape at this volume range (Table 7.2 and 7.3). As the bubble turns into a teardrop shape, the extent decreases. As soon as, the bubble shape starts growing in the horizontal direction, i.e., when it finally reaches the cap shape, the extent starts to increase. This increase is more significant for the 0.25 wt% concentration compared to the 0.40 wt% concentration due its shape not

reaching cap shape. For 0.18 wt% PAAM concentration, there is not enough data to observe the changes described above.



Figure 7.5. Shape parameter, extent as a function of the bubble volume for different concentrations of PAAM solution.

Next, the solidity of the bubbles is investigated. The solidity shows how concave a bubble is. A value of 1 for the solidity means that the bubble has a convex shape. It can be seen from Figure 7.6 that the solidity is independent of the polymer concentration. The solidity, first increases, then around 200 mm³ bubble volume, it starts decreasing. It is predicted that as the cusp at the bubble tail becomes more pronounced at this volume range, the bubble becomes more concave (see Table 7.2 and 7.3).

The third parameter is the circularity of the bubble. If the circularity is 1, it means that the shape is perfectly circular. First of all, it is necessary to state that the circularity is drawn for bubble volumes larger than 25 mm³. The circularity measurement could not be performed for the bubbles that are smaller than 25 mm³ since the code could not measure the circularity parameter correctly. When a bubble is very small, the edge of the bubble is not clear enough to detect due to the low resolution.

Table 7.2. Parameters and dimensionless numbers for 0.25 wt% PAAM solution.

Bubble	V	d	UB	a/b	a	b	С	Ext	Sol	E-1	E-2	β	Ý	Re	Bo	We	Ca	CD
0	4	2	5	1.9	2.07	1.11	-	0.78	0.984	0.94	0.98	0.65	2.66	0.04	0.55	8.E-04	0.1	914
0	12	3	37	1.7	2.80	1.64	-	0.77	0.987	0.88	1.01	0.63	13.14	0.87	1.07	5.E-02	0.4	27
0	51	5	163	1.9	4.53	2.41	0.99	0.75	0.989	0.94	1.01	0.57	35.50	10.74	2.79	2.E+00	1.8	2
	100	6	207	1.6	5.20	3.17	0.99	0.75	0.990	0.82	1.11	-	35.89	17.24	4.41	3.E+00	2.3	2
	305	8	242	1.0	5.46	5.38	0.83	0.74	0.988	0.51	1.53	-	28.94	25.91	9.25	7.E+00	2.7	2
	490	10	231	-	-	-	0.69	0.73	0.985	0.41	1.77	-	23.63	25.93	12.69	7.E+00	2.6	2
	663	11	236	-	-	-	0.61	0.76	-	0.32	2.11	-	21.84	28.05	15.52	8.E+00	2.7	3
	2012	16	264	-	-	-	-	-	-	-	-	-		-	-	1	-	-

Bubble	v	d	UB	a/b	а	b	C	Ext	Sol	E-1	E-2	β	Ϋ́	Re	Bo	We	Ca	CD
0	5	2	2	1.8	2.17	1.18	-	0.78	0.984	0.93	0.99	0.73	0.75	0.00	0.61	8.E-05	0.2	10699
0	12	3	11	1.7	2.80	1.67	-	0.76	0.986	0.87	1.01	0.62	3.92	0.06	1.07	5.E-03	1.1	299
	50	5	70	1.6	4.52	2.81	1.00	0.74	0.990	0.85	1.02	0.56	15.19	1.37	2.79	3.E-01	6.7	12
	106	6	78	1.5	5.75	3.77	0.98	0.73	0.991	0.82	1.02	0.53	13.25	1.79	4.56	5.E-01	7.5	13
	301	8	196	-	-	-	0.96	0.70	0.990	-	1.02	0.48	23.55	9.37	9.17	4.E+00	18.8	3

Table 7.3. Parameters and dimensionless numbers for 0.40 wt% PAAM solution (part 1/2).

Bubble	V	d	UB	a/b	a	b	С	Ext	Sol	E-1	E-2	β	Ý	Re	Bo	We	Ca	CD
	496	10	239	-	-	-	0.94	0.70	0.985	-	1.12	-	24.33	13.81	12.79	8.E+00	23.0	2
	661	11	251	-	-	-	0.92	0.68	0.980	-	1.09	-	23.19	15.43	15.49	9.E+00	24.1	2
*	2027	16	252	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-



Figure 7.6. Shape parameter, solidity as a function of the bubble volume for different concentrations of PAAM solution.

The circularity is almost 1 for 25 mm³ for all concentrations. At this stage the bubbles are changing from the spherical to the slightly prolate shape. Then, it starts to decrease slowly until a volume of 100 mm³. From 25 to 100 mm³, the circularity of the bubble in 0.40 wt% PAAM solution is less than the circularity observed in other concentrations. Around 100 mm³, the circularity abruptly decreases for the bubbles of 0.18 and and 0.25 wt% PAAM concentrations. It is clear from Table 7.2 that the shape suddenly starts growing in the horizontal axis and deviates from the circularity. However, the circularity of the bubble in 0.40 wt% concentration is still close to 1 until 300 mm³ and then it deviates significantly but less steeply than the circularity observed for 0.18 and 0.25 wt% solutions. The circularity change of 300 to 500 mm³ bubble volume for the 0.40 wt% PAAM solution can be seen in Table 7.3.

The next shape parameters are a, b and their ratio, a/b. As explained in Section 7.10, a is the minor axis and the smallest distance of the bubble and b is the distance between the center of gravity to the tip of the bubble. Figure 7.8 shows that a does not depend on the concentration until ~100 mm³. For larger bubble volumes, a is constant for 0.18 and 0.25 wt% concentrations where it keeps increasing for the 0.40



Figure 7.7. Shape parameter, circularity as a function of the bubble volume for different concentrations of PAAM solution.

wt% concentration until ~1000 mm³. It should be noted that as the parameter a is defined as the minimum distance value for a bubble, when the bubble starts growing horizontally, at some point the definitions of a and b are switched (see Figure 5.2, which change for wide bubbles). This switch is seen at around 100 mm³ bubbles for 0.18 and 0.25 wt% PAAM solutions, and at around 500 mm³ bubbles for 0..40 wt% PAAM solution (see Table 7.3).

The major axis, b increases constantly for all concentrations as seen in Figure 7.9. At these critical values mentioned in the previous paragraph, b becomes the longest of bubble volumes distance in the horizontal direction. In parallel with these results, the a/b value, as shown in Figure 7.10, it first decreases at different rates for different concentrations as the bubble turns from spherical to prolate and from prolate to teardrop shape, then increases as a starts growing much faster in the horizontal direction. Finally, after the aforementioned critical values, a/b starts decreasing.

The eccentricity-1, E1, defines the ratio of the length of a bubble's short (minor) axis to the length of its long (major) axis. This parameter shows how elliptical a



Figure 7.8. Shape parameter, minor axis, a as a function of the bubble volume for different concentrations of PAAM solution.



Figure 7.9. Shape parameter, b parameter as a function of the bubble volume for different concentrations of PAAM solution.



Figure 7.10. Shape parameter, ratio of the minor axis to the major axis, a/b as a function of the bubble volume for different concentrations of PAAM solution.

bubble is. For this parameter, the same pattern is observed with a/b as their definitions are similar to each other. However, sometimes a/b sometimes E1 is preferred in the literature. Hence, here both are presented. It is observed that the eccentricity of the bubble decreases as the major axis increases. For example, the bubble becomes less eccentric as the volume changes from 5 to 80 mm³ for the 0.40 wt% concentration. Then it starts to increase due to the horizontal expansion of the bubble and finally decreases around 400-500 mm³ due to the definition of major and minor axis (Figure 7.11). Also, one can say from Figure 7.11 that the bubble with a volume of ~300 mm³ is more eccentric for 0.25 wt% than 0.40 wt%. Bubbles of both 0.25 wt% and 0.40 wt% concentrations become significantly eccentric after 80 mm³ and 400 mm³, respectively (see Table 7.3). Finally, the difference in the eccentricity becomes negligible at very high volumes where the bubbles of both concentrations have a cap shape.

The next parameter is the eccentricity-2, E2, and is defined as the ratio of the equivalent diameter to the minor axis. It should be reminded that the minor axis is in the horizontal direction for the small volumes, and then it shifts to the vertical direction since the bubbles grow much faster horizontally. E2 being close to 1 shows



Figure 7.11. Shape parameter, eccentricity-1, E1, as a function the bubble volume for different concentrations of PAAM solution.

that the bubble has a circular shape. Unlike the previous parameters, the equivalent diameter is obtained through the volume. It can be seen from Figure 7.12 that E2 is independent of the PAAM concentration until ~80 mm³ bubble volume. E2 is constant at around 1, i.e., bubbles with almost circular shape, until some volume. Then, there is deviation from the circularity, i.e., the eccentricity increases. This increase happens at lower bubble volumes in solutions with lower concentrations. The E2 for bubbles of volumes around 500-600 mm³ in 0.25 wt% PAAM concentration is almost double the E2 for bubbles in 0.40 wt% PAAM solution. Thus, it can be concluded that the bubbles are axysymmetrical.

The next parameter is β , which is defined by Xu *et al.* (2019) [37] to determine the bubble volume at which a cusp is formed at the tail of the bubbles rising in the PAAM and xanthan gum solutions (Figure 7.13). β is an empirical parameter and applied to one side of the bubble only as the bubbles are assumed to be axisymmetric. If β is less than 0.60 for a bubble, then it should have a cusp according to Xu *et al.* (2019) [37]. This empirical correlation is applied to this system and the results are given for a determination coefficient, R^2 of 0.9. The results are presented in Figure



Figure 7.12. Shape parameter, eccentricity-2, *E*2, as a function of the bubble volume for different concentrations of PAAM solution.

7.13 where the red line shows $\beta = 0.6$.



Figure 7.13. Shape parameter, β as a function of the bubble volume for different concentrations of PAAM solution.

The results show that β can be determined from the fits for a bubble volume range of 10-200 mm³. The results are more representative and in accordance with

visual observations of the cusp for the 0.40 wt% concentration. The graphs shows that a bubble that has a volume larger than ~25 mm³ should have a cusp for all concentrations. For this purpose, a bubble volume range of 15-30 mm³ is specifically studied. The findings are in a good match with the study of Xu *et al.* (2019) as the first bubbles with a cusp are observed at 26 mm³, 24 mm³ and 24 mm³ for 0.18 wt%, 0.25 wt%, 0.40 wt% PAAM concentration studies, respectively (see Figure 7.14). Xu *et al.* (2019) detected the cusp formation ($\beta < 0.6$) for the xanthan gum solutions, but not for the PAAM concentrations of 0.10, 0.25, 0.50, 1.00 and 1.25 wt%.

As mentioned earlier at the beginning of this section while discussing the discontinuity detection for different PAAM polymers, it is seen that similar results are obtained for the same types and brands of the polymer. Here, we report a cusp formation for the same bubble volume range for the 0.25 wt% PAAM (Sigma Aldrich) solution in this study where Xu *et al.* (2019) did not observe any. Here, we also conclude that the cusp formation volume is concentration independent.

Concentration (wt%)	0.18	0.25	0.40
Volume (mm ³)	26	24	24
	Q		0

Figure 7.14. The bubbles where the cusp is seen for the first time at $\beta \approx 0.59$ for different PAAM concentrations.

Finally, for the bubble dynamics of the PAAM solutions, dimensionless numbers are calculated. The definitions of the dimensionless numbers are given in Section 5.1.3. First, the Bond number, Bo, is presented versus the Reynolds number, Re, in Figure 7.15. Bo represents the ratio of the gravitational forces over the surface tension forces, and Re is the ratio of the inertial forces to the viscous forces. Bo is constant for small

Re and it is less than 1. Then, Bo increases suddenly at different values of Re for each concentration.

At first, Re increases due to the increase in the equivalent diameter, d and the terminal velocity, u_B . In the mean time, Bo increases only with d. For a larger bubble volume, u_B becomes constant as shown in Figure 7.4 and the only contribution to Re comes from d which includes the viscosity behaviour index, n (n < 1 for shear-thinning liquids) as its power. While the increase in Re slows down due to this, Bo increases proportional to the square of d which results in an abrupt increase. Thus, the effect of the surface tension decreases and the bubble shape deviates from the spherical to the cap shape (see Tables 7.2 and 7.3). Also, each concentration shows a sudden increase in Bo at different Re since the polymer concentration changes the rheological properties, n and K.



Figure 7.15. Bo versus Re for different concentrations of PAAM solution.

The next dimensionless number is the Capillary number, Ca, that is the ratio of the viscous forces to the surface tension forces in the system (see Figure 7.16). The behaviour is similar to what is observed in Re vs. Bo in Figure 7.15. For a given Re,
Ca of 0.40 wt% concentration is much higher than the other concentrations due to the high viscosity of 0.40 wt% solution.



Figure 7.16. Ca versus Re for different concentrations of PAAM solution.

Next, Weber number, We, which represents the ratio of the inertial forces to the surface tension forces is plotted in Figure 7.17. For a given equivalent diameter, the inertial forces are greater than the surface tension forces. The increase in We is smaller than the increase in Bo. The effect of the polymer concentration shows its effect on We at small d. As d increases, u_B reaches the plateau value for all concentrations and the only contribution to We comes from d.

Finally, the drag coefficient, C_D , is calculated according to Eq. 5.12 as plotted in Figure 7.18. $C_{D,2}$ and $C_{D,3}$ show the drag coefficient for Hadamard-Rybczynski regime and are valid for Re <1 and Re <60, respectively; and it is applicable for deformable surfaces. It is observed that C_D is slightly higher than $C_{D,2}$ at different Re regions for each concentration. Figure 7.18 shows that the bubbles act like solid spheres at $Re \approx$ 0.001 - 0.002 for 0.40 wt%, at $Re \approx 0.02 - 0.2$ for 0.25 wt% and at $Re \approx 0.3 - 1.0$ for 0.18 wt% (see the first bubble of Tables 7.2 and 7.3). For these regions, the bubbles are



Figure 7.17. We versus Re for different concentrations of PAAM solution.

deviate from Hadamard-Rybczynski regime, to the C_D of Stokes' law which is 24/Re and not shown on the plot. A transition from Stokes to Hadamard-Rybczynski regime occurs where these given Re regions end.

For larger volumes, the bubbles start fitting Hadamard-Rybczynski regime after the given Re regions for each concentration. For Re=1, the results fit $C_{D,3}$ regime until Re=60 which is in a good correlation with the definition of $C_{D,3}$ [32,91].

In the literature, it is reported that the discontinuity at the bubble velocity is observed at the transition from the Stokes to the Hadamard-Rybczynski regime [9]. It was also reported that the cusp starts forming at this transition [30]. However, as stated before, there is no discontinuity in the bubble velocity at a critical volume for this system. Additionally, the cusp formation does not occur at these transitions for any of the polymer concentrations. As stated previously, the cusp forms around 25 mm³, and the *Re* value is much higher for these bubble volumes.



Figure 7.18. C_D versus Re for different concentrations of PAAM solution.

7.1.3. Effect of the Injection Period

The injection period is a crucial part of the bubble dynamic studies as it is proved to affect the results considerably. The injection period depends on the physical properties of the polymer used and also the bubble size.

To determine the injection period that does not affect the results, an experiment was designed specifically for this purpose. It is made sure that only a single bubble rises throughout the column. The tested injection periods for a bubble volume of 2000 mm 3 are tabulated in Table 7.4

The results show that there exists a critical injection period for each concentration. The injection periods of 15, 33 and 57 s were determined to be safe for 0.10, 0.25 and 0.40 wt% concentrations, respectively. The increase in the injection period with the increasing polymer concentration is in an agreement with literature [19]. Below these injection periods, it is determined that the terminal bubble velocity, u_B increases and reaches a plateau value for the entire bubble volume range. The results also shows

	$0.10 \mathrm{~wt\%}$	$0.25 \mathrm{wt\%}$	$0.40 \mathrm{~wt\%}$
	0.5	4.5	11
Doriod (a)	5	9	25
Period (S)	15	20	32
	-	33	57

Table 7.4. Tested injection periods for different PAAM

that the bubble shapes are also affected with decreasing injection period. The bubble shapes elongate in the vertical direction. The increase in the bubble terminal velocity and the elongated bubble shape were also confirmed by Funfschilling and Li (2006) for 0.5 wt% PAAM (913 SH, SNF Floerger) solution [21]. In the light of this investigation, the bubble injection period is set to 60 s for a bubble volume range of $\sim 10 - 2000 \text{ mm}^3$ in this study for all PAAM concentrations.

7.1.4. Effect of the of Surfactant on Bubble Dynamics

Surfactants have an extremely important role in the dynamics of the bubbles as they alter the surface tension forces. For this purpose, the polymer, PAAM concentration is kept constant at 0.25 wt% and different concentrations of SDS (a water soluble surfactant) are added. The surface tension of 0.25 wt% PAAM solution is 74 mN/m as shown in Section 4.5. The surface tension is lowered to 66.8 and 45.2 mN/m for 10 and 100 ppm of SDS, respectively. 10 ppm of SDS is shown not to have any significant effect on bubble dynamics.

First of all, the average bubble terminal velocity, u_B is studied and shown in Figure 7.19. The results show that 10 ppm SDS slightly affects the terminal velocity until ~50 mm³. When we look at the effect of 100 ppm, it can be seen that the terminal velocity significantly decreases for a given bubble volume. However its effect diminishes around ~250 mm³ and the bubble terminal velocity for all concentrations reaches a plateau value at ~250 mm³. The decrease in the bubble velocity with the addition of the surfactant is expected [1] and can be explained by the Marangoni effects. Tables 7.5 and 7.6 show the bubble images and respective bubble dynamics data of 0.25 wt% PAAM solution with 10 ppm and 100 ppm, respectively. In these tables, V, d, u_B , a, b, C, Ext, Sol, E-1, E-1, E-2 are volume (mm³), equivalent diameter (mm), the minor axis (mm), the distance from center of gravity to the tail (mm), circularity, extent, solidity, eccentricity 1 and 2, respectively.



Figure 7.19. The average terminal velocity as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.

Extent results of the bubbles are given in Figure 7.20. The surfactant concentration does not have any effect on the extent for bubble volumes up to 30 mm³ and the ratio of the bubble area to its bounding box decreases similarly. In this regime, the bubbles have a prolate shape and evolve into the teardrop shape. The bubble area decreases compared to the bounding box. The bubbles start expanding in the horizontal direction for the PAAM solution around \sim 30 mm³. However, the extent value decreases for the PAAM solutions with the surfactant in it. It means that the bubbles are not growing horizontally as much as the bubbles do in clean liquids, without surfactant. Solutions with 10 ppm SDS show that the decrease in the extent slows down after \sim 50 mm³.

Bubble	V	d	UB	a/b	a	b	С	Ext	Sol	E-1	E-2	β	Ý	Re	Bo	We	Ca	CD
0	4	2	5	1.9	1.96	1.05	•	0.79	0.984	0.94	0.99	0.69	2.82	0.04	0.55	9.E-04	0.1	847
0	20	3	44	1.6	3.33	2.07		0.74	0.987	0.85	1.01	0.56	13.01	1.22	1.67	1.E-01	0.5	23
0	49	5	160	1.8	4.55	2.48	-	0.74	0.989	0.95	1.00	0.60	35.19	10.39	3.04	2.E+00	2.0	2
•	104	6	218	1.6	5.27	3.25	0.97	0.72	0.987	0.81	1.11	0.23	37.43	18.77	4.99	4.E+00	2.7	2
0	190	7	239	1.4	5.76	4.25	0.91	0.71	0.982	0.68	1.24	0.43	33.54	23.70	7.47	6.E+00	3.0	2

Table 7.5. Parameters and dimensionless numbers for 0.25 wt% PAAM solution with 10 ppm SDS.

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Bubble	V	d	UB	a/b	a	b	С	Ext	Sol	E-1	E-2	β	Ϋ́	Re	Bo	We	Ca	CD
0	5	2	2	1.9	2.18	1.17	-	0.76	0.981	0.94	0.97	0.65	1.17	0.01	0.97	3.E-04	0.0	4479
0	18	3	17	1.7	3.18	1.92	÷	0.76	0.987	0.85	1.02	0.62	5.30	0.28	2.28	2.E-02	0.3	144
Q	53	5	47	1.3	4.48	3.42	0.93	0.70	0.987	0.73	1.04	0.43	10.07	1.56	4.72	2.E-01	0.9	28
	100	6	129	1.4	5.68	4.19	0.92	0.68	0.981	0.77	1.01	0.28	22.40	8.25	7.19	2.E+00	2.4	5
	195	7	214	1.8	6.94	3.93	0.91	0.67	0.974	0.88	1.04	0.33	29.68	19.95	11.24	7.E+00	3.9	2

Table 7.6. Parameters and dimensionless numbers for 0.25 wt % PAAM solution with 100 ppm SDS.

Table 7.5 shows that the bubble expands horizontally around this volume whereas this is not observed for a bubble in the presence of 100 ppm SDS and consequently, the bubbles have elongated shape. For the larger bubbles, the extent parameter keeps decreasing for 100 ppm since the bubbles are still vertically elongated whereas for the solution without surfactant, the extent increases due to the cap formation.



Figure 7.20. Shape parameter, extent as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.

The next parameter is the effect of the solidity and the results are given in Figure 7.21. In Section 7.1.2, it was shown that the solidity is independent of the polymer concentration in the absence of the surfactant. The solidity increases for all of the solutions and becomes closer to 1, i.e., the bubble reaches a convex shape. This can be explained by the bubbles turning from a spherical to a prolate shape. However, the solidity changes with the addition of the surfactant for a given volume after ~100 mm³ bubble volume. The decrease is significant for both 10 and 100 ppm surfactant concentration. When Tables 7.2, 7.5 and 7.6 are checked simultaneously for a bubble volume of around 100 mm³, one can see that the concavity increases at the lower half of the bubble as the surfactant concentration increases. As the surfactant

concentration increases at the rear of the bubbles (tail), the surface tension forces are weakened and the bubbles stretch out vertically.



Figure 7.21. Shape parameter, solidity as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.

The effect of the surfactant concentration on the circularity of the bubble is given in Figure 7.22. The circularity decreases quickly for the PAAM solution with 100 ppm surfactant whereas the decrease is slower for the other concentrations. Similar to the concavity, the elongation caused by the weakened surface tension forces makes the bubble deviate from its circular shape. However, the circularity curves cross each other around $\sim 200 \text{ mm}^3$. After this point, the bubble grows in the horizontal direction and deviates from the circularity for the solution that has no surfactant.

Next, a, b and a/b parameters are presented in Figures 7.23, 7.24 and 7.25, respectively. The results show that the surfactant does not have any effect on the minor axis, a. The change for the 0.25 wt% PAAM solution without the surfactant after ~200 mm³ is caused by the change of the axes and explained earlier in Section 7.1.2. Figure 7.24 shows that the surfactant does not have any effect on b for spherical and prolate bubbles, i.e., bubbles of small volumes. However, the increase in the b



Figure 7.22. Shape parameter, circularity as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.

is much faster for the 100 ppm concentration compared to the other solutions. As explained in this section for the earlier parameters, the tail of the bubbles elongates. Therefore, the distance between the center of mass and the tip of the bubble increases. The a/b clearly shows that the bubbles also start growing horizontally (after ~50 mm³). This change can be observed from Table 7.6 for the bubbles with the volumes of 53 and 100 mm³. This behaviour is also similar to the effect of increasing PAAM concentration on a/b.

When the eccentricity-1, E1 is studied, it is observed from Figure 7.26 that the surfactant does not affect this parameter until $\sim 20 \text{ mm}^3$. However, the tail elongates and becomes less eccentric with increasing bubble volume in the presence of 100 ppm surfactant.

The observations for the eccentricity-2, E2 Figure 7.27), are similar to the ones presented for PAAM solutions in the absence of the surfactant. 10 ppm surfactant does not have any effect, and the eccentricity remains around 1 for the 100 ppm surfactant



Figure 7.23. Shape parameter, minor axis, a as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.



Figure 7.24. Shape parameter, b as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.



Figure 7.25. Shape parameter, the ratio of minor axis, a over b as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.

concentration while others are increasing. It can be seen from Tables 7.5 and 7.6 that the bubble (195 mm^3) is more circular for a solution with 100 ppm SDS than the bubble (190 mm^3) in a solution with 10 ppm SDS.

Lastly, β is studied to understand the effect of the surfactant on the cusp formation and is shown in Figure 7.28. It is reported in Section 7.1.2 that the cusp formation occurs around 25 mm³ for 0.25 wt% in the absence of the surfactant. The cusp formation starts at a slightly lower volume once the surfactant is added. The cusp becomes more pronounced with the increase of the surfactant which can also be observed from the bubble images from Table 7.5 (49 mm³) and Table 7.6 (53 mm³). This observation is in a good agreement with the work of Li *et al.* (2011) who reported that cusps become more pronounced as the surfactant concentration increases for different concentrations of PAAM solutions with Triton X-100 as the surfactant [97]. Previously, we concluded that the cusp formation is concentration independent whereas it is highly dependent on the surfactant concentration.



Figure 7.26. Shape parameter, eccentricity, E1 as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.



Figure 7.27. Shape parameter, eccentricity, E2 as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.



Figure 7.28. Shape parameter, β as a function of the bubble volume for 0.25 wt% PAAM solution with different surfactant concentrations.

First dimensionless number studied is the Bond number, Bo for different concentrations of the surfactant. The results are given in Figure 7.29 and show that 10 ppm of surfactant has almost no effect on the system. However, 100 ppm surfactant SDS significantly increases Bo for a given Re, except at high Re. Since the surfactant decreases the surface tension, the increase in Bo is expected. One should remember that for a given Re, bubbles may have different volumes as the terminal velocity decreases with the surfactant concentration. For example, it can be seen from Tables 7.5 and 7.6 that 195 mm³ bubble rising in a solution with 100 ppm SDS has a higher Bo (11.24) than the 190 mm³ bubble in a solution with 10 ppm SDS (7.47). Therefore, the change detected in Figure 7.29 is actually the effect of the surfactant on the terminal velocity observed in Re.

Next, the capillary number, Ca is presented in Figure 7.30. For a given Re, Ca is higher with the surfactant for a certain range of Re. For a given bubble diameter, the changes that come from the terminal velocity and the surface tension will compete in Ca (Eq. 5.10). Once, Tables 7.5 and 7.6 are studied together, it is found that



Figure 7.29. Bo versus Re for different concentrations of surfactant in PAAM solution.

for the small bubbles the terminal velocity is dominating, but as the bubble diameter increases, the contribution of surface tension is more pronounced.

The next dimensionless number is the Weber number, We (Figure 7.31). Similar to Ca, the terminal velocity and the surface tension compete for a given bubble diameter as the surfactant concentration changes (Eq. 5.11). However, We is proportional to the square of the terminal velocity. As the bubble diameter increases, the effect of the surfactant on the terminal velocity diminishes. For small bubble volumes, without surfactant We is large since the velocity is high which contributes to We more than the surface tension.

Lastly, C_D is studied to examine the effect of the surface tension on the drag. It is found that C_D increases and does not fit Hadamard–Rybczynski regime for Re < 1 $(C_{D,2})$. When the surfactant is added into the solution, it accumulates on the bubble surface which causes bubble act like a solid sphere. The increase in the C_D with the addition of the surfactant is also observed in the literature and explained by the



Figure 7.30. Ca versus Re for different concentrations of surfactant in PAAM solution.



Figure 7.31. We versus Re for different concentrations of surfactant in PAAM solution.

decrease in the bubble terminal velocity [63,97]. Therefore, C_D increases, and it starts deviating from the Hadamard–Rybczynski towards the Stokes' regime for small bubble volumes. The results show that C_D fits the Hadamard–Rybczynski regime for Re < 60, $C_{D,3}$ which means the accumulation on the bubble surface is not too high to deviate C_D from the Hadamard–Rybczynski regime.



Figure 7.32. Drag coefficient, C_D versus Re for different concentrations of surfactant in PAAM solution.

7.1.5. The Bubble Trajectory

When a bubble rises in a liquid column, it tends to have an axi-asymmetric trajectory such as zigzag or spiral, depending on the physical characteristics and the bubble volume of the liquid. Therefore, we also studied the trajectory of the rising bubble. In this section, 3 different solutions are studied, namely 0.25 wt% PAAM, 0.40 wt% PAAM and 0.25 wt% PAAM with 100 ppm SDS solutions. A bubble volume range of 20 to 2000 mm³ is presented here. The x-axis shows the horizontal deviation of the bubble in mm where the y-axis represents the tube length/equivalent diameter.

Here, it should be reminded that the bubble terminal velocity is lowered when

the surfactant is added into the system. In this kind of a system, the small bubbles $(20 \text{ and } 50 \text{ mm}^3)$ are recorded for a fixed number of frames. Therefore, the bubbles for this system are recorded for a limited time due to the low terminal velocity.

Figures 7.33, 7.34 and 7.35 show the trajectory results for bubbles in 0.25 wt% PAAM, 0.40 wt% 40 PAAM and 0.25 wt% PAAM with 100 ppm SDS solutions, respectively. The results show that the bubbles first deviate to the right most of the time which can be due to an experimental bias. Later, it seems like the bubbles turn to the core of the column. As the polymer and the surfactant concentrations increase, the path instability and the bubble oscillation increase and the bubble do a lateral motion, which is in agreement with the literature [65, 98, 99].



Figure 7.33. The trajectory of bubbles with different volumes in 0.25 wt% PAAM solution.

It should be noted that the deviation grows in the x-direction as the bubble volume increases. However, it reaches a maximum of 0.45 mm where the equivalent diameter of the biggest bubble represented here is 7 mm for 2000 mm³. Also, a bubble can be recorded for a maximum of 6 cm after leaving the inlet completely in the vertical direction.



Figure 7.34. The trajectory of bubbles with different volumes in 0.40 wt% PAAM solution.



Figure 7.35. The trajectory of bubbles with different volumes in 0.25 wt% PAAM with 100 ppm SDS solution.

7.1.6. Unsteady-state Bubble Dynamics

In this section, the evolutions of the bubbles' volumes, shapes and velocities while rising through the liquid are given. The unsteady results are presented as a function of the tube length (the vertical distance travelled by the bubble)/equivalent diameter. Here the bubble volume, the bubble velocity and an example shape parameter are given for 0.25 wt% PAAM, 0.40 wt% PAAM and 0.25 PAAM with 100 ppm SDS solutions.

In Figure 7.36, the evolution of the bubble volume for 0.25 wt% PAAM solution. The results show that the bubble volume reaches steady state except the highest bubble volume, 1985 mm³. When a similar bubble volume range is studied for 0.40 wt% PAAM solution (Figure 7.37), a similar observation is done. However, the oscillation in the largest bubble volume (2026 mm³) decreases as the polymer concentration increases. A smaller range of bubble volume is studied for 0.25 wt% in the presence of 100 ppm SDS, given in Figure 7.38. Hence, no such unsteady behavior is observed.



Figure 7.36. The evolution of the bubble volume as a function of tube length/equivalent diameter for 0.25 wt% PAAM.



Figure 7.37. The evolution of the bubble volume as a function of tube length/equivalent diameter for 0.40 wt% PAAM.



Figure 7.38. The evolution of the bubble volume as a function of tube length/equivalent diameter for 0.25 wt% PAAM with 100 ppm SDS.

The next parameter studied is the bubble velocity and given in Figures 7.39, 7.40 and 7.41 for three different concentrations. Similarly, an ongoing oscillation occurs for 1985 mm³ in Figure 7.39 due to the wobbling of the bubble. Therefore, this size of bubble should not be considered in steady-state analysis. Especially, when SDS is added, the oscillation amplitude is larger. When Figures 7.40 and 7.41 are studied, it is seen that it takes longer time for the velocity to reach the steady-state compared to the volume.

Finally, the shape parameter, minor axis, a, is chosen as an example and presented in Figures 7.42, 7.43 and 7.44 for 0.25 wt% PAAM, 0.40 wt% PAAM and 0.25 wt PAAM with SDS solutions, respectively. It is observed that there occurs small changes at the beginning of the motion, then the bubbles reach a steady-state value for the rest of the distance travelled. Again, 1985 mm³ in Figure 7.42 and 2026 mm³ in Figure 7.43 show oscillations for the recorded distance. The rest of the bubbles presented in Figures 7.42, 7.43 and 7.44 reach a steady-state value quickly. The largest bubbles of both Tables 7.2 and 7.3 show oscillation and marked with a sign, *; and their bubble dynamics results are not given. Also, the step changes (for example 50 mm³ in Figure 7.43) are observed due the replacement of a with the shape parameter, b as the bubble expands horizontally.

The results show that it is necessary to do an unsteady-state pre-analysis to determine the safe bubble volume range to present steady results for the chosen solution and the distance recorded. For an unsteady-state analysis, it is important to observe the motion of a bubble for a long distance (tube length).



Figure 7.39. The evolution of the bubble velocity as a function of tube length/equivalent diameter for 0.25 wt% PAAM.



Figure 7.40. The evolution of the bubble velocity as a function of tube length/equivalent diameter for 0.40 wt% PAAM.



Figure 7.41. The evolution of the bubble velocity as a function of tube length/equivalent diameter for 0.25 wt% PAAM with 100 ppm SDS.



Figure 7.42. The evolution of the shape parameter, a as a function of tube length/equivalent diameter for 0.25 wt% PAAM.



Figure 7.43. The evolution of the shape parameter, a as a function of tube length/equivalent diameter for 0.40 wt% PAAM.



Figure 7.44. The evolution of the shape parameter, a as a function of tube length/equivalent diameter for 0.25 wt% PAAM with 100 ppm SDS.

7.2. Birefringence Measurements and Bubble Dynamics for Different PAAM Concentrations with 0.40 wt% HPC

7.2.1. Bubble Dynamics for Different PAAM Concentrations with 0.40 wt% HPC

The stress measurement around the rising bubble could not be obtained in polyacrylamide (PAAM) solutions as PAAM is not birefringent enough. Hence, hydroxy propyl cellulose (HPC) is added. In this section, HPC concentration is kept constant at 0.40 wt%. Therefore, the legends of the graphs do not include the concentration information to avoid crowded legends.

When the bubble dynamics of the PAAM solutions are studied, it is observed that bubbles in 0.10 wt% PAAM showed a 3D motion. However, when HPC is added to the same solution, bubbles exhibit axysymmetric motion. It should be noted that when 0.40 wt% HPC is added to the 0.40 wt% PAAM solution, the solution becomes extremely viscous and is not employed in this study.

First, the average bubble terminal velocity is presented in Figure 7.45. The bubble volume range is between 20 and 3500 mm³ for 0.10 and 0.18 wt% PAAM solutions, whereas the range starts from 100 mm³ for 0.25 wt%. Similar to the only PAAM solutions, there is no sign of velocity jump at some critical bubble volume. Similarly, in all solutions, the terminal velocity versus the bubble volume reaches a plateau value. The average bubble terminal velocity in a HPC added solution is lower than that in only PAAM solution.

The first shape parameter is the extent which shows the ratio of the area of the bubble to the area of its bounding box. It can be seen from Figure 7.46 that until 150 mm^3 , this parameter is concentration independent. However, the decrease in the extent value slows down for 0.10 wt% PAAM solution where the decrease is much faster for the other solutions. Consequently, the extent for bubbles larger than 150 mm³ in a

0.10 wt% PAAM solution, is smaller than for bubbles in other solutions. The bubble becomes more concave as the polymer concentration increases which can also be seen from Tables 7.7, 7.8 and 7.9. Also, a bubble with a volume of around 100 mm³ in a HPC added 0.25 wt% PAAM solution, is more concave. One can attribute this change to the increase in the viscoelasticity of the solution.



Figure 7.45. The average terminal velocity as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.

Next, the shape parameter, solidity is studied in Figure 7.47. The solidity directly measures the concavity of a bubble. Previously, it was reported that this parameter is independent of the polymer concentration (Figure 7.6) but is significantly affected by the addition of the surfactant into the solution. Figure 7.47 shows that the solidity is again concentration independent. The rapid decrease in the solidity (increase in the concavity) is clear in Table 7.7 as the volume increases from 748 to 1000 mm³.

When there is no HPC in the PAAM solution (Figure 7.7), the circularity is constant around 1 until 150 mm³ bubble volume. However, once HPC is added, the circularity decreases starting from a bubble volume of 70 mm³ (Figure 7.48). The bubbles of the lowest PAAM concentration are close to the circular shape. Table

Bubble	V	d	UB	a/b	a	b	С	Ext	Sol	E-1	E-2	β	Ϋ́	Re	Bo	We	Ca	CD
0	31	4	34	1.6	3.85	2.34	-	0.75	0.986	0.86	1.01	-	8.62	0.72	2.01	6.E-02	1.2	45
0	104	6	99	1.6	5.81	3.69	0.97	0.72	0.989	0.85	1.00	-	16.92	4.21	4.51	8.E-01	3.4	8
0	197	7	141	1.6	7.22	4.47	0.96	0.71	0.990	0.89	1.00	-	19.49	7.90	6.91	2.E+00	4.9	5
0	498	10	217	1.5	8.46	5.70	0.93	0.71	0.984	0.74	1.16	-	22.02	17.44	12.82	6.E+00	7.5	3
0	748	11	226	1.3	8.92	6.83	0.88	0.70	0.977	0.65	1.26	-	20.03	19.97	16.81	8.E+00	7.8	3

Table 7.7. Parameters and dimensionless numbers for 0.10 wt% PAAM + 0.4 wt% HPC (part 1/2).

Table 7.7.	Parameters and	dimensionless	numbers for	0.10 wt%	PAAM -	⊢ 0.4 wt%	HPC (part 2/	(2).
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Bubble	v	d	UB	a/b	a	b	С	Ext	Sol	E-1	E-2	β	Ϋ́	Re	Bo	We	Ca	Съ
	1004	12	235	1.1	8.68	8.01	0.81	0.70	0.967	0.54	1.43	-	18.90	22.37	20.46	9.E+00	8.1	3
	1523	14	237	0.8	8.48	9.99	0.64	0.69	0.934	0.43	1.68	-	16.63	24.62	27.01	1.E+01	8.2	3
	2007	16	243	0.8	8.70	11.28	0.61	0.68	0.930	0.39	1.80	-	15.52	26.81	32.47	1.E+01	8.4	3

Bubble	v	d	UB	a/b	а	b	С	Ext	Sol	E-1	E-2	β	Ý	Re	Bo	We	Ca	CD
0	30	4	11	1.6	3.77	2.39	-	0.75	0.986	0.82	1.02	-	2.76	0.08	1.97	6.E-03	0.9	445
0	106	6	51	1.3	5.57	4.25	0.93	0.71	0.989	0.72	1.06	-	8.75	1.01	4.58	2.E-01	4.6	29
0	207	7	94	1.4	7.08	5.23	0.93	0.70	0.989	0.75	1.04	-	12.84	2.83	7.14	9.E-01	8.4	11
0	496	10	177	1.4	9.66	6.67	0.91	0.67	0.979	0.82	1.02	-	17.98	8.47	12.79	4.E+00	15.7	4
Q	751	11	202	1.5	11.01	7.19	0.91	0.67	0.972	0.87	1.02	-	17.88	11.07	16.85	6.E+00	18.0	4

Table 7.8. Parameters and dimensionless numbers for 0.18 wt% PAAM + 0.4 wt% HPC (part 1/2).

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Bubble	V	d	UB	a/b	a	b	С	Ext	Sol	E-1	E-2	β	Ϋ́	Re	Bo	We	Ca	CD
0	981	12	224	1.7	11.47	6.83	0.91	0.67	0.971	0.84	1.07	-	18.16	13.55	20.14	8.E+00	20.0	3
0	1495	14	244	-	-	-	0.89	0.66	0.963	-	1.11	-	17.18	16.49	26.69	1.E+01	21.7	3
6	2065	16	249	-	-	-	0.80	0.66	0.952	-	1.32	-	15.78	17.97	33.09	1.E+01	22.2	3

Table 7.8. Parameters and dimensionless numbers for 0.18 wt% PAAM + 0.4 wt% HPC (part 2/2).

Bubble	v	d	UB	a/b	а	b	С	Ext	Sol	E-1	E-2	β	Ϋ́	Re	Bo	We	Ca	CD
0	116	6	33	1.3	5.64	4.43	0.91	0.72	0.982	0.69	1.07	-	5.51	0.28	4.86	9.E-02	8.5	71
0	206	7	53	1.2	6.84	5.50	0.91	0.71	0.989	0.69	1.07		7.19	0.62	7.12	3.E-01	13.4	35
0	505	10	119	1.2	9.42	8.09	0.85	0.66	0.979	0.67	1.05	-	12.02	2.56	12.95	2.E+00	30.2	9
0	750	11	159	1.3	11.11	8.56	0.85	0.66	0.974	0.75	1.01	-	14.15	4.32	16.84	4.E+00	40.6	6

Table 7.9. Parameters and dimensionless numbers for 0.25 wt% PAAM + 0.4 wt% HPC (part 1/2).

Bubble	V	d	UB	a/b	a	b	C	Ext	Sol	E-1	E-2	β	Ý	Re	Bo	We	Ca	CD
0	998	12	189	1.4	12.30	9.04	0.85	0.66	0.966	0.79	1.01	-	15.27	5.90	20.38	6.E+00	48.2	5
	1548	14	244	1.7	13.19	7.97	0.92	0.68	0.978	0.83	1.09	-	16.99	9.37	27.30	1.E+01	62.1	3
	2016	16	249	-	-	-	0.86	0.65	0.951	-	1.06	-	15.89	10.04	32.57	1.E+01	63.4	3

Table 7.9. Parameters and dimensionless numbers for 0.25 wt% PAAM + 0.4 wt% HPC (part 2/2).



Figure 7.46. Shape parameter, extent as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.



Figure 7.47. Shape parameter, solidity as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.

7.7 shows that the bubbles of 10 wt% PAAM solution rapidly extend horizontally after reaching a critical volume, which is 550 mm³. Tables 7.8 and 7.9 show that the bubbles are first elongated vertically until the critical volume, then they start expanding horizontally and become closer to a circular shape until very high bubble volumes (> 1000 mm³). This oscillation in the circularity is not observed for the PAAM solutions in the absence of HPC nor when a surfactant is added.



Figure 7.48. Shape parameter, circularity as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.

Next, a, b and a/b shape parameters are investigated in Figures 7.49, 7.50 and 7.51. It is mentioned in Sections 7.1.2 and 7.1.4 that as the bubble sizes grow horizontally, a and b are swapped. Figure 7.49 shows that the a is independent of the polymer concentration until a bubble volume of around 1000 mm³. Afterwards, a and b are swapped for bubbles in 0.10 and 0.18 wt% PAAM. A similar behaviour is observed for b (Figure 7.50). When the plot of a/b is analyzed in Figure 7.51, one can say that bubbles grow vertically first, then horizontally. Similar observations can be done for the eccentricity-1, E1 (Figure 7.52) as for a/b parameter. The bubbles show a horizontally expanding eccentric (elliptic) shape for volumes higher than 100, 200 and 325 mm³ rising in 0.10, 0.18 and 0.25 wt% PAAM solutions with HPC, respectively.



Figure 7.49. Shape parameter, minor axis, a as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.



Figure 7.50. Shape parameter, b as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.


Figure 7.51. Shape parameter, the ratio of the minor axis to b, a/b as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.



Figure 7.52. Shape parameter, eccentricity-1, E1 as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.

The shape parameter, eccentricity-2 informs about the circularity. Figure 7.53 shows that as the concentration of the polymer increases the bubble deviates more from the circular shape and become more elliptic. Tables 7.7, 7.8 and 7.9 also show largest bubbles in the 0.10 wt% PAAM solution are more elliptic. These observations with the circularity shape parameter results verify that the bubbles are axysymmetric.



Figure 7.53. Shape parameter, eccentricity-2, E2 as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.

The last shape parameter for this section is β which shows the presence of a cusp at the tip of the bubble. In Sections 7.1.2 and 7.1.4, it is shown that this parameter works well for a certain bubble volume range and predicts the critical bubble volume successfully. The shape parameter, β results are presented in Figure 7.54. It is found that the β value is below 0.6 which indicates that a cusp should be formed for the entire bubble volume range. However, the determination coefficient, R^2 is highly below 0.9. Therefore, one can say that this parameter does not work when HPC is added into the PAAM solutions. The bubble shapes can be seen from Tables 7.7, 7.8 and 7.9 where there is a cusp formation. One should remember that this parameter is obtained empirically for only PAAM solutions [37]. Adding 0.40 wt% HPC, which may be responsible for this incompatibility, changes the rheological properties.



Figure 7.54. Shape parameter, β as a function of the bubble volume for different concentrations of PAAM with 0.40 wt% HPC solution.

The first dimensionless number studied in this section is the *Bo* number and given in Figure 7.55. For a constant PAAM concentration, when the rheological properties of the solutions are compared (Table 3.4), it can be seen that the flow consistency index, *K* increases highly with the addition of HPC. Also, for a given bubble diameter, the terminal velocity decreases. Both of these changes cause a decrease in *Re* for a given bubble diameter. For a given bubble volume, *Bo* is constant ($Bo \approx 13$ for 490 mm³ in Table 7.2 and 505 mm³ in Table 7.9). Therefore, we can say that this plot is shifted to the left when HPC is added into the PAAM solution at constant concentration.

Next, Ca is studied in Figure 7.56. The behaviour is similar to that without HPC as presented in Figure 7.16. The terminal velocity of the bubbles decrease and the viscosity of the solution increases as HPC is added. The contribution of the viscosity is dominant; therefore, Ca increases.

The behavior of the Weber, We, versus Re (Figure 7.57) is similar to what is obtained without HPC (Figure 7.17). We becomes smaller when HPC is added into



Figure 7.55. Bo versus Re for different concentrations of PAAM with 0.40 wt% HPC.



Figure 7.56. Ca versus Re for different concentrations of PAAM with 0.40 wt% HPC.

the solution for the 0.25 wt% PAAM. Hence, the inertial forces decrease when HPC is added. However, when the bubble size increases (V $\approx 2000 \text{ mm}^3$), We stays constant with the HPC addition. Since the bubble terminal velocity reaches the same plateau value, its effect diminishes and both solutions have the same We.



Figure 7.57. We versus Re for different concentrations of PAAM with 0.40 wt% HPC Bo against Re for different concentrations of PAAM solution.

Finally, C_D is presented in Figure 7.58. In the presence of HPC, only 0.25 wt% PAAM is close to the Hadamard-Rybczynski regime for Re < 1, and it fits $C_{D,3}$ for Re > 1. However, the other solutions do not fit these regimes. The increase in the C_D can be attributed to the surface of the bubble becoming more rigid in the presence of HPC.

7.2.2. Stress Measurement Through Birefringence

In this section, the results of the stress measurement around a rising bubble will be presented and discussed. The details about the method, the experimental setup and how the stress is quantified are given in Sections 2.3, 4.1 and 5.2, respectively.



Figure 7.58. C_D versus Re for different concentrations of PAAM with 0.40 wt% HPC.

In the literature, for some work the presence of the discontinuity is attributed to the formation of a negative wake at the trailing tip of the bubble, or to the cusp formation. On the other hand, some work argue that the discontinuity in the velocity and the cusp formation are not related.

In the previous section (Section 7.2.1), we report that there is no discontinuity at the velocity for the polymer, the concentration and the bubble volume range investigated. Therefore, in this section, the focus is on the stress around the bubble and to explain the relationship between the stress and any of the above quantities, i.e., the cusp formation and the velocity discontinuity, if there is any.

The results are given in terms of the retardance, δ and the stretching angle, χ . Birefringence, Δn is directly proportional to the retardance with a constant (see Eqn. 5.18). Therefore, it is not given here separately. Both δ and χ are given for a bubble volume range of 140-1285 mm³ at three different PAAM concentrations (0.10, 0.18 and 0.25 wt%) in the presence of a constant HPC concentration of 0.40 wt% are presented. The retardance measurements are presented in Figures 7.59, 7.62 and 7.66 for increasing bubble volume where each column represents a polymer concentration increasing from left to right. Same representation is valid for the stretching angle measurements in Figures 7.60, 7.63 and 7.67. The intensity scales are given on the right side of each row.

The measurements are performed only for the left half of the bubbles as they are assumed axisymmetric. In the previous section, this assumption was proved by analyzing the circularity and eccentricity-2, E2 results together. As the light is deemed due to the polarizers, the recording is not of high resolution. All the cutting of the bubbles into half is automated; however, some are not very clear due to low resolution and the difficulty in edge detection.

When Figure 7.59 is analyzed, the retardance intensity is not high for the given bubble volume range. However, it can still be noticed that the retardance increases from low to high concentration and for a constant polymer concentration, a slight increase can be seen with the growing bubble size. Figure 7.60 also shows a similar pattern, i.e., the polymer stretches more with increasing polymer concentration and the bubble volume. At this bubble volume range, the bubbles have prolate shape and are convex (Figure 7.61). Also, bubbles rising in 10 wt% PAAM solution has the highest average terminal velocity while having the lowest retardance around it (Figure 7.2.2).

As the bubble volume increases, a significant change immediately appears in both δ and χ as given in Figures 7.62 and 7.63, respectively. It is observed that the stress occurs on top of the bubble due to the squeezing and also around the lower half of the bubbles. The stretching angle, χ clearly shows the effect of the increase in the bubble volume for a given bubble concentration in Figure 7.63.



Figure 7.59. The retardance, δ measurement for different PAAM concentrations for a bubble volume range of 140-238 $\rm mm^3.$



Figure 7.60. The stretching angle, χ measurement for different PAAM concentrations for a bubble volume range of 140-238 $\rm mm^3.$



Figure 7.61. Shape parameter, solidity as a function the bubble volume for the bubbles studied in the stress measurement.



In smaller volumes (Figure 7.59), it is observed that the retardance, δ is significantly less for low concentration solutions. However, when a larger volume range (360 to 600 mm³) is considered in Figure 7.62. it is observed that the δ abruptly increases for the lowest polymer concentration. Figure 7.2.2 shows that the terminal velocities of large bubbles in all concentrations are very close to each other. However, a significant difference in δ exists at the tail.

The effect of the bubble shape through bubble parameters is investigated on the retardance, δ . Figure 7.61 shows that the bubbles have similar concavity for all concentrations until 1000 mm³ bubble volume. When the shape parameters, a and a/b are studied in Figures 7.64 and 7.65, it is seen that the minor axis, a is replaced with the shape parameter b as the bubble is highly expanded horizontally, especially for high concentrations. Thus, the sudden increase in the retardance can be linked to the change in the bubble shape. Figure 7.63 shows that the stretching angle, χ is dissipated to a wider region as the PAAM concentration increases. The change in the bubble shape for a constant volume can be connected to the dissipation of the intensity.

When the bubble volume range increases to 750-1285 mm³, it is observed that the retardance, δ (Figure 7.66) significantly increases on the top of the bubble. It is also noticed that the stretching angle, χ (Figure 7.67) increases at the trailing tip of the bubbles. The increase in both δ and stretching angle, χ intensities may be reflecting the negative wake and the cusp formation.

It can be concluded from the birefringence experiment that the stress around a rising bubble is directly in a relationship with the bubble shape. As the bubble volume increases, this relationship becomes more prominent. Even tough, we could not characterize the cusp formation with the empirical shape parameter, β due to the addition of HPC, the bubbles showed a cusp formation (see Tables 7.7, 7.8 and 7.9). We proved that the cusp formation and the stress occurring at the tip of the bubble are not prerequisites for the the discontinuity at the bubble terminal velocity.



Figure 7.62. The retardance, δ measurement for different PAAM concentrations for a bubble volume range of 360-600 $\rm mm^3.$



Figure 7.63. The stretching angle, χ measurement for different PAAM concentrations for a bubble volume range of 360-600 $\rm mm^3.$



Figure 7.64. Shape parameter, minor axis, a as a function the bubble volume for the bubbles studied in the stress measurement.



Figure 7.65. Shape parameter, the ratio of the minor axis to b, a/b as a function the bubble volume for the bubbles studied in the stress measurement.



Figure 7.66. The retardance, δ measurement for different PAAM concentrations for a bubble volume range of 750-1285 $\rm mm^3.$



Figure 7.67. The stretching angle, χ measurement for different PAAM concentrations for a bubble volume range of 750-1285 mm³.

7.3. Non-Isothermal Experiments

In Section 7.1, the bubble dynamics is presented for an isothermal system. In this section, the bubbles rising PAAM solution is studied in the presence of a temperature gradient applied in the horizontal direction.

The experimental setup for this system is explained in Section 4.4. A temperature gradient is formed by applying heat from one wall whereas the other wall is kept constant at 25°C using a water bath. Temperature gradients of 25-40°C and 25-50°C are created and the experiments are performed when the system reaches the steady-state. 0.18, 0.25 and 0.40 wt% PAAM solutions are tested.

Even though no measurable effect is observed in any of the solutions in terms of bubble terminal velocity and bubble shape, the average terminal velocity and circularity results are presented for 0.18 wt% (Figures 7.68, 7.69) and 0.40 wt% (Figures 7.70, 7.71) PAAM solutions.

A temperature gradient is observed through Schlieren mehtod, especially on the tail of a bubble rising in the solution, as presented in Figure 7.72. Apparently this gradient is not enough to cause an observable effect on the bubble dynamics. If a larger temperature gradient is applied, there is a risk of polymer degradation. So, one method may be to apply the temperature gradient along the vertical direction.



Figure 7.68. The average terminal velocity as a function of the bubble volume for 0.18 wt% PAAM solution: isothermal (maroon) and 15 °C temperature gradient (pink).



Figure 7.69. Shape parameter, circularity as a function of the bubble volume for 0.18 wt% PAAM solution: isothermal (maroon) and 15 °C temperature gradient (pink).



Figure 7.70. The average terminal velocity as a function of the bubble volume for 0.40 wt% PAAM solution: isothermal (black) and 15°C temperature gradient (purple).



Figure 7.71. Shape parameter, circularity as a function of the bubble volume for 0.40 wt% PAAM solution: isothermal (black) and 15°C temperature gradient (purple).



Figure 7.72. The temperature gradient observed at the tails of a small and a big bubble.

8. CONCLUSIONS AND RECOMMENDATIONS

In this chapter, the dissertation is concluded by summarizing the most important and relevant results of the study and recommendations are provided for future studies.

8.1. Conclusions

In this dissertation, the dynamics of a bubble rising in a viscoelastic liquid is studied. In the literature, there are various unanswered questions regarding especially a jump in the steady bubble velocity that occurs at a critical bubble volume. Some studies attribute the discontinuity to the negative wake and the cusp formation at the tail of the bubble; whereas others claim that there is no relationship amongst these parameters. Therefore, a study is designed to answer these questions or find evidences to understand the behavior of a rising bubble in a large bubble volume range.

The bubble dynamics is studied using different concentrations of PAAM solutions, which show both viscoelastic and shear-thinning rheology. For different PAAM concentrations, it is observed that the terminal velocity reaches a plateau with increasing bubble volume in accordance with the literature. We did not obtain any evidences of the presence of a discontinuity at the bubble terminal velocity for the studied bubble volume range and polymer concentrations. The detailed literature survey regarding the PAAM types and brands shows that the discontinuity is directly related to the manufacturing of the polymer. In the literature, there is no experiment with the PAAM used in this study (AN 934 SH by SNF Floerger) showed a discontinuity in the bubble velocity.

Bubble shapes observed with increasing bubble volume are spherical, prolate, teardrop with a cusp, and cap-shape. Shape parameters included in this study are extent, solidity, circularity, minor axis, a, the distance between the center of gravity and tip of the bubble tail, b, the a/b ratio, eccentricity-1, eccentricity -2, and the empirical shape parameter, β . The results show that the solidity and b are concentrationindependent parameters. Solidity showed how a bubble reaches a more concave shape with increasing bubble volume. Additionally, extent shows the change of bubble shape from prolate to teardrop. Circularity, a, and eccentricities show the effect of the polymer concentration on the bubble shape. A bubble loses its circularity, becomes wider horizontally, and more elliptic after a critical bubble volume for low polymer concentration. The empirical cusp parameter, β developed for PAAM solutions is successfully used to determine the critical bubble volume for the cusp formation. In this dissertation, the results show that the critical bubble volume is independent of the polymer concentration. In the literature, it was claimed that the cusp formation causes a discontinuity. Here, we show that there is no discontinuity in the bubble velocity in the presence of a cusp.

The dimensionless numbers, Re, Bo, Ca, We, and C_D are analyzed to understand the forces acting on the system for a broad range of bubble volume and different polymer concentrations. For smaller bubble volumes, the concentration is important for Re and We due to its effect on the bubble velocity. As the bubble volume increases for a given PAAM concentration, gravitational forces become dominant for Bo, and the effect of the surface tension decreases which causes a change in the bubble shape from spherical to cap-shape. For low concentrations and Re <1, the spherical-shaped bubbles act like a solid sphere and deviate from C_D of Hadamard Rybczynski which is for deformable surfaces. For higher Re, larger bubble volumes fit Hadamard Rybczynski regardless of the polymer concentration. Even though a transition occurs from Stokes to Hadamard Rybczynski, there is no velocity discontinuity at a critical bubble volume, as suggested by some work in the literature.

Since the surface tension forces play an important role in the bubble dynamics as seen from *Bo* versus Re in the PAAM studies, the effect of the surfactant is studied for 0, 10 and 100 ppm concentrations. 10 and 40% decrease occurred in the surface tension for 10 and 100 ppm SDS concentrations, respectively. The study shows that 10 ppm is effective only for the extent and the solidity parameters and has a negligible effect on the velocity and the other shape parameters. As the surfactant concentration increases, the bubbles slow down. However, the average terminal velocity reaches the same plateau value regardless of the surfactant concentration. The bubble expands less in the horizontal direction and elongates more in the vertical direction in the presence of the surfactant. Overall, the bubbles are less distorted in the presence of the surfactant. There exist critical volumes for the surfactant to affect the concavity and the circularity of bubbles. It is shown that the cusp formation occurs for smaller bubble volumes when a surfactant is added into the system and is highly dependent on the surfactant concentration whereas the polymer concentration did not have any effect.

The effect of the surfactant concentration on Re and Ca diminishes due to the plateau regime as the bubble volume increases. However, its effect becomes more prominent for Bo. It is also found that even for small surfactant concentration, drag increases and deviates from Hadamard Rybczynski regime. This is explained by the accumulation of the surfactant on the bubble surface where Hadamard Rybczynski applies to deformable surfaces only. It fits Hadamard Rybczynski equation for higher Re.

For the PAAM studies, the effect of the injection period is studied to determine a safe period that does not affect the results and found that the required injection period increases as the polymer concentration increases; and 60 s is chosen as the injection period for the experiments which is in a good agreement with the literature.

In this study, the bubble velocity and shape parameters are mostly presented at steady-state. However, the unsteady analysis of the bubbles is also important to determine whether steady-state is reached. It is shown that the bubbles do a wobbling motion as the polymer concentration decreases. Also for a given polymer concentration, wobbling occurs above a critical volume. The bubble trajectory is studied and it is found that the bubbles most of the time move towards the right side of the column which is believed to be experimentally biased. The increase in the polymer and surfactant concentration increases the path instability and the oscillation of the rising bubble. However, the lateral movement of the bubble is found to be very small.

Finally, the stress field around the bubble as it rises in the viscoelastic liquid is studied. The stress-optic law, i.e., the flow birefringence method is used and the retardance of the light beam occurs in the presence of the stress. HPC is added into the PAAM solution to give the birefringent property. In the literature, some work hypothesized that the negative-wake (downwards flow) and the cusp are pre-requisites for the discontinuity at the bubble velocity, and some reported they are not related. Here, it is shown that a discontinuity did not exist even though a negative-wake and cusp start forming. A negative-wake is observed at a critical bubble volume, which is much higher than the critical discontinuity volumes reported in the literature. For smaller bubble volumes, the polymer concentration has a significant effect on retardance. However, it is found that the bubble shape becomes important for stress as the bubble volume increases. Therefore, the stress around a rising bubble is directly related to the bubble shape.

At last, the bubble dynamics is studied in the presence of a temperature gradient. Even though a temperature gradient is successfully achieved, no significant change is observed in the bubble dynamics. Schlieren method showed that the convection was not clearly visible.

In the beginning of this study, it is hypothesized that there could be a critical bubble volume where the bubble's shape and the average terminal velocity, the polymer concentration and the stress around the rising bubble are correlated. It is successfully shown which shape parameters and dimensionless numbers are important for different cases such as the cusp formation or stress around the bubble for a wide bubble volume range, and various polymer or the surfactant concentrations. It is proven that the shape parameters are as important as the polymer concentration for the stress formation around the rising bubble above a critical bubble volume. This study also emphasizes the importance of the rheology in the bubble dynamics in non-Newtonian liquids. The uncertainty in the literature concerning the discontinuity in the terminal velocity of the bubble at a critical volume, the cusp formation, the stress around the bubble and the negative wake can be enlightened if only the bubble dynamics studies are integrated with the rheological properties of the liquids, especially the viscoelasticity.

8.2. Recommendations

The present work contributes to the bubble dynamics in viscoelastic liquids by studying multiple parameters. This field is still full of unknowns and requires more controlled and focus work to have a better understanding of the bubble motion.

When our findings are combined with a detailed literature survey, it is found that there is likely to be a relationship between the discontinuity and PAAM used; and the studies are suffering from a lack of information and a detailed rheology analysis. Since polymers can differ from one batch to another and cover a broad range of molecular weight, it is suggested to employ custom design polymers with known rheological properties. This would help to contribute to the literature by minimizing the unknowns related to the polymer and giving specific and reproducible results that can be used to compare and improve.

In the bubble dynamics, the experiments are performed for a broad range of bubble volumes which creates difficulty for the analysis since the light, camera, and recording settings are sensitive. Therefore, the study should be divided into smaller sections of the bubble volume to get more accurate results. Besides, the field of view of the camera needs to be increased without sacrificing the resolution for the unsteady-state and the trajectory studies. On the other hand, with this big number of data, principle component analysis may give a correlation between different shape parameters.

Similar arguments are also valid for surfactant experiments. Additionally, wider range of concentration and surfactant type could be employed. The injection system for the bubbles needs to be improved and automatized as it mostly depends on the experimentalist in the current system and the syringe pump can create a limited volume range. Considering the discontinuities reported in the literature occur at very small volumes, the effect of the experimentalist should be removed.

The edge detection consists of an important part of the birefringence study. Another camera is used to measure the bubble volume, then the recordings of two videos are matched later which can cause errors. Even though the birefringence results are highly promising, it would benefit from a high-speed camera with high resolution, and the entire analysis can be automated. Also, during the experiments, the birefringence method is found to be promising to study the bubble breakage and coalescence; and the wall-effect. Thus, the study can be extended to a new direction.

Finally, a detailed understanding is required for the non-isothermal experiments before designing a new experimental setup. The results clearly show that the current system is not appropriate to understand the effect of the temperature gradient. Our studies with the Schlieren method showed that the study of the effect of the temperature gradient in the vertical direction is promising Applying the birefringence method to the non-isothermal study would also be interesting.

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APPENDIX A: UNCERTAINTY ANALYSES

If a quantity is not determined explicitly in an experiment and is based on the variables that are obtained directly through the experiments, an uncertainty analysis is required. Uncertainty analysis offers us a methodological approach to the sensitivity of the results. With this approach, a range for possible errors is determined. In this section, the uncertainty analyses done for the length, the bubble velocity and the volume are given.

A.1. Uncertainty Analysis for the Length

The measured pixel length of the calibration object and the uncertainty from the MATLAB's *imdistline* function is x = 818.6 pixel ± 1.4 pixel ($\Delta x=1.4$ pixel). The length and the uncertainty of the calibration object is $y=76.2 \pm 0.1$ mm ($\Delta y=0.1$ mm). The length of a pixel, p and its uncertainty value, Δp in mm are calculated as

$$p = \frac{y}{x},\tag{A.1}$$

where Δp is

$$\Delta p = \sqrt{\left(\frac{dp}{dy}\right)^2 (\Delta y)^2 + \left(\frac{dp}{dx}\right)^2 (\Delta x)^2}.$$
 (A.2)

The uncertainty in pixel becomes

$$\Delta p = \sqrt{\frac{1}{x^2} (\Delta y)^2 + \frac{y^2}{x^4} (\Delta x)^2}.$$
 (A.3)

Thus, the pixel length with its uncertainty, **p** becomes 0.0931 ± 0.0002 mm/pixel using Eq. A.1-A.3.

A sample uncertainty analysis for the minor axis, a of a bubble is shown below where a=85 pixel ($\Delta a=1$ pixel). The length of a in millimeter is m and its uncertainty, Δm are 7.9 mm and 0.1 mm through Eq. A.4-A.6, respectively.

The length of a in millimeter is calculated as

$$\mathbf{m} = \mathbf{p} \times \mathbf{a},\tag{A.4}$$

where the uncertainty becomes

$$\Delta m = \sqrt{\left(\frac{dm}{dp}\right)^2 (\Delta p)^2 + \left(\frac{dm}{da}\right)^2 (\Delta a)^2}$$
(A.5)

and

$$\Delta m = \sqrt{a^2 (\Delta p)^2 + p^2 (\Delta a)^2}.$$
 (A.6)

The minor axis in millimeter with its uncertainty, **m** becomes 7.9 ± 0.1 mm

A.2. Uncertainty Analysis for the Velocity

The uncertainty analysis is done as follows where u, Δu , z and t are the bubble velocity, the uncertainty in the bubble velocity, the path that the bubble travels and the time difference between two consecutive video frames, respectively.

The uncertainty in the velocity is

$$\Delta \mathbf{u} = \sqrt{\left(\frac{\mathrm{d}\mathbf{u}}{\mathrm{d}\mathbf{z}}\right)^2 (\Delta \mathbf{z})^2 + \left(\frac{\mathrm{d}\mathbf{u}}{\mathrm{d}\mathbf{t}}\right)^2 (\Delta \mathbf{t})^2},\tag{A.7}$$

where it turns into the following in order as

$$\Delta u = \sqrt{\frac{1}{t^2} (\Delta z)^2 + \frac{z^2}{t^4} (\Delta t)^2},$$
 (A.8)

$$\frac{\Delta u}{u} = \frac{t}{z} \sqrt{\frac{1}{t^2} (\Delta z)^2 + \frac{z^2}{t^4} (\Delta t)^2}, \qquad (A.9)$$

$$\frac{\Delta u}{u} = \sqrt{\left(\frac{\Delta z}{z}\right)^2 + \left(\frac{\Delta t}{t}\right)^2}.$$
 (A.10)

A sample uncertainty analysis for the bubble velocity, u is done where the pixel length of the path that the bubble's center of gravity takes between the two consecutive frames, z_{pixel} is 15 pixel \pm 1 pixel.

The distance in millimeters of the bubble's center of gravity between two frames, z is calculated as

$$z = p \times z_{pixel} . \tag{A.11}$$

The uncertainty in the distance in millimeters of the bubble's center of gravity becomes

$$\Delta z = \sqrt{\left(\frac{dz}{dp}\right)^2 (\Delta p)^2 + \left(\frac{dz}{dz_{pixel}}\right)^2 (\Delta z_{pixel})^2},$$
(A.12)

and

$$\Delta z = \sqrt{\left(z_{\text{pixel}}\right)^2 \left(\Delta p\right)^2 + p^2 \left(\Delta z_{\text{pixel}}\right)^2}.$$
(A.13)

Thus, z and Δz are calculated as 1.4 mm and 0.1 mm, respectively.

Finally, u and Δu are determined as 140 mm/s and 10 mm/s, respectively, where $t = 0.0100 \pm 0.0001$ s. Therefore, the uncertainty analysis for the velocity becomes $\mathbf{u} = 140 \pm 10$ mm/s.

A.3. Uncertainty Analysis for the Volume

The truncated-cone method is used through Eq. 5.2 to calculate the bubble volume. R and r are found by the edge detection of the bubble; and both of their uncertainties are 1 pixel ($\Delta R=1$ pixel and $\Delta r=1$ pixel).

Hence, the uncertainty in the truncated-cone volume, ΔV_i is

$$\Delta V_{i} = \sqrt{\left(\frac{dV_{i}}{dr}\right)^{2} (\Delta r)^{2} + \left(\frac{dV_{i}}{dR}\right)^{2} (\Delta R)^{2}}, \qquad (A.14)$$

and

$$\Delta V_{i} = \frac{\pi h}{3} \sqrt{(2r+R)^{2} (\Delta r)^{2} + (2R+r)^{2} (\Delta R)^{2}}.$$
 (A.15)

The bubble volume, V is

$$\mathbf{V} = \sum_{i=1}^{N} \mathbf{V}_i,\tag{A.16}$$

where the uncertainty in the bubble volume ΔV is calculated as follows

$$\Delta V = \sqrt{(\Delta V_1)^2 + (\Delta V_2)^2 + (\Delta V_3)^2 + \dots + (\Delta V_N)^2}.$$
 (A.17)

The calculated volume and the uncertainty of the volume are found in pixel³ unit and the following formulas is applied to calculate the volume as

$$V_{mm^3} = p^3 \times V_{piksel^3}, \tag{A.18}$$

where its uncertainty in mm³ is calculated as follows

$$\Delta V_{mm^3} = \sqrt{\left(\frac{dV_{mm^3}}{dp}\right)^2 (\Delta p)^2 + \left(\frac{dV_{mm^3}}{dV_{piksel^3}}\right)^2 \left(\Delta V_{piksel^3}\right)^2}, \qquad (A.19)$$

and

$$\Delta V_{mm^3} = \sqrt{\left(3p^2 x V_{piksel^3}\right)^2 (\Delta p)^2 + (p^3)^2 \left(\Delta V_{piksel^3}\right)^2}.$$
 (A.20)

The volume and the uncertainty of a sample bubble are found as $V_{pixel^3} = 910000 \text{ pixel}^3$ and $\Delta V_{piksel^3} = 2200 \text{ piksel}^3$. Finally, the bubble volume, **V** with its uncertainty is calculated as $V_{mm^3} = 730 \text{ mm}^3 \pm 5 \text{ mm}^3$

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Expected presentation date	Jun 2021	
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