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CHARACTERIZATION OF BUBBLE SHAPES IN NON-NEWTONIAN FLUIDS BY PARAMETRIC EQUATIONS

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Abstract

Based on the experiment of single air bubbles rising in stagnant non-Newtonian fluids, an innovative model containing the aspect ratio (E) and two parameters (α , β) was proposed and proved to be capable of characterizing the bubble shape from spherical/ellipsoidal to prolate/oblate-tear with good accuracy. Several impacts on bubble deformation were investigated involving the rheological properties of the fluids and different forces exerted on the bubble which were quantified by multiple dimensionless numbers (e.g. Reynolds number, Eötvös Number, Deborah number). Within a wide range ($-9 < \log Mo < 3$, Mo : Morton number), the empirical correlations were obtained for parameter β , and between α and β . Together with the shape model, a complete system was set up for bubble shape characterization and prediction that will provide new ideas for future studies on bubble hydrodynamics.

Keywords: Bubble shape, Bubble dynamics, Gas-liquid system, Non-Newtonian fluid

1 Introduction

Bubble-liquid systems are widespread in industries such as oil extraction, wastewater treatment and bioreactors. For these applications, the knowledge of bubble shape is of importance since it is directly related to hydrodynamic behavior (e.g. terminal velocity, drag, surface area) and can influence the mixing level and the efficiency of energy, mass, and momentum transfer.

The observed shapes of individual bubbles in free motion in Newtonian fluids have been well investigated and can be divided mainly into three categories: spherical, ellipsoidal, and spherical-cap or ellipsoidal-cap [1,2]. Grace et al. [3] have proposed a generalized graphical correlation or the so-called "Grace diagram" that delineates the shapes of bubbles in Newtonian fluids, which provides a guideline for the further studies on the bubble shape [4–6].

On the other hand, although numerous studies deal with bubbles rising in non-Newtonian media, only a few of them have discussed and analyzed the bubble shape [7–12]. It is generally agreed that non-Newtonian fluids have complex rheological properties (e.g. shear thinning, viscoelasticity, etc.) leading to a variety of bubble shapes, such as teardrop or elongate bubbles [13–18].

To quantitatively characterize the bubble shape, many researchers used the aspect ratio or eccentricity i.e. the ratio between the lengths of the major and minor axes of the bubbles. Pioneering work by Moore [19] and Tadaki and Maeda [20] theoretically correlated the aspect ratio of the bubble with the Weber number and original Tadaki number ($Ta = Re.Mo^{0.23}$), respectively. Their correlation were modified and improved for small inertial effects [21] and became applicable to clean and contaminated bubbles [22–24] for a large range of Morton numbers [25]. More recently, many researchers have begun to realize that, for fluids with high viscosity, no single dimensionless number (Eo , We , or Ta number) is suitable to predict the bubble deformation. Therefore, more dimensionless numbers have been used and many empirical models have been built to fit the results of the new experiments or numerical simulations [25–28]. For instance, Liu et al. [29] suggested that, in highly viscous fluids, the bubble shape is dramatically affected by the viscosity and the effects of We should be considered along with Re . However, the aspect ratio suffers from the shortcoming of being unable to characterize the details of shape or irregular forms (e.g. a cap bubble and an ellipsoidal bubble would have the same aspect ratio). A few researchers have tried to use other parameters to characterize the bubble deformation. Myint et al. [30] introduced a distortion factor to quantify the asymmetry between the upper and lower parts of the bubble divided by the major axis. Another study, by Yan and Zhao [31], used the bubble circularity, defined as the ratio of the circumference of an equivalent circle to that of the real bubble projection. Although some valuable conclusions have been drawn from the previous work, most of the expressions mentioned above could only handle quasi-ellipsoidal bubbles. For a bubble with a more complicated contour (e.g. cap, teardrop), it is clear that neither the aspect ratio nor the circularity can properly characterize the bubble shape.

Therefore, in this paper, a new model using the parametric equations is proposed to directly depict the complete profile of the bubble. Experiments will be implemented to study individual air bubbles of different sizes (equivalent diameters: 0.7-7 mm) rising in stagnant non-Newtonian fluids (aqueous solution of polyacrylamide or Xanthan gum) by means of high-speed photography and image post-processing technologies. The objective mainly aims to investigate the overall impact (including the different dimensionless numbers and the rheological properties of the fluid) on the bubble shape.

2 Materials and method

2.1 Experimental setup

The experimental setup for studying bubble shapes is shown in **Figure 1**. A column (volume: $100 \times 100 \times 300 \text{ mm}^3$) was filled with 2 L of the liquid under study. A single bubble was generated by injecting the air into the column through a horizontal nozzle located 20 mm above the column bottom by a syringe pump (Harvard Apparatus, PHD 22/2000) with the flow rate of $10 \text{ } \mu\text{L/h}$. Stainless steel nozzles of different calibers ($\approx 0.5\text{-}1 \text{ mm}$) were used to make bubbles of different sizes (equivalent diameters: $0.7\text{-}7 \text{ mm}$). A high-speed camera (Photon SA3, resolution: 1024×1024 pixels) was placed next to one side of the column and focused on the rising bubble with a vertical image area ($\approx 40 \times 40 \text{ mm}^2$). The recording rate of the bubble images was 2000 frames per second. The image area was illuminated by a backlight board placed at the opposite side of the column. The images acquired by the camera were transmitted to the computer and processed with a professional software (PFV-Photon FASTCAM Viewer). All the experiments were performed at 293.15 K and under atmospheric pressure.

Figure 1. Experimental setup for bubble shape investigation.

2.2 Materials and the rheological properties

Two typical non-Newtonian solutions that are widely used in industries [32] were chosen for the experiments: poly(acrylamide-co-acrylic acid) partial sodium salt (PAAm) (Sigma-Aldrich, CAS: 62649-23-4) and Xanthan gum (Sigma-Aldrich, CAS: 11138-66-2). The solutions were prepared by dissolving powder of substances in deionized water, under five concentrations for PAAm (0.1%, 0.25 %, 0.5%, 1%, 1.25 % wt.) and three for Xanthan gum (0.1 %, 0.25 %, 0.5% wt.).

Figure 2. Flow curves (viscosity versus shear rate) for PAAm solutions and Xanthan gum solutions

The rheological properties of both PAAm and Xanthan solutions were measured by a rheometer (HAKKE MARS III, Germany) at 293.15 K with the shear rates ranging from 0.001 s⁻¹ to 10000 s⁻¹. The measured flow curves are plotted in Figure 2. It is found that Xanthan solutions have much higher viscosity than that of the PAAm solutions, especially at low shear rates (<10 s⁻¹). Meanwhile, both of the two solutions present a similar flow behavior. Under low and high shear rates, the viscosity approaches a Newtonian plateau where the value of viscosity trends to be constant. Between these two plateaus, the viscosity decreases with higher shear rate which is known as the shear-thinning property. This evolution can be characterized by the classic Carreau model [33,34]:

$$\frac{\mu - \mu_{\infty}}{\mu_0 - \mu_{\infty}} = [1 + (\lambda \dot{\gamma})^2]^{\frac{n-1}{2}} \quad (1)$$

where:

- μ is the variable viscosity [Pa·s];
- μ_0 is the viscosity at the zero shear rate [Pa·s];
- μ_{∞} is the viscosity at the infinite shear rate [Pa·s];
- $\dot{\gamma}$ is the variable shear rate [s⁻¹];
- λ is the relaxation time [s];
- n is the power index.

For each solution under different concentrations, multiple samples were tested and fitted with the Carreau model. The fitting results are in agreement with the experimental ones with the determination coefficient $r^2 > 97\%$, proving that the Carreau model is suitable for describing the viscosity variation of the investigated non-Newtonian fluids in our cases. The averaged results of the regression of the viscosity are shown in Table 1 as well as other general physical properties. In this study, the shear rate $\dot{\gamma}$ near the bubble was estimated by $\dot{\gamma} = U/D_{eq}$ with the experimental data: bubble rising velocity U and equivalent diameter D_{eq} . This

is the characteristic shear rate near the equator of a spherical bubble. The effective shear rate may be slightly different for large, non-spherical bubbles, but for simplicity we use the same definition in all cases [35]. The methods for calculating U and D_{eq} are presented in **Section 2.3** and **Section 2.4.1**.

Table 1. Materials and their physical properties ($T = 293.15$ K; the viscosity is valid for the shear rate ranging between 0.001 s^{-1} and 1000 s^{-1}).

Composition	Concentration [wt.]	σ [mN/m]	ρ [kg/m ³]	μ_0 [Pa·s]	μ_∞ [Pa·s]	λ [s]	n
PAAM	0.1%	69	998	0.109	0.003	5.91	0.38
	0.25%	67	999	0.115	0.004	6.77	0.40
	0.5%	66	1000	0.120	0.005	8.19	0.42
	1%	65	1003	0.146	0.007	9.18	0.42
	1.25%	65	1004	0.159	0.008	11.86	0.43
Xanthan	0.1%	69	998	0.36	0.001	15.12	0.47
	0.25%	67	999	3.6	0.002	26.11	0.33
	0.5%	66	1001	69	0.004	59.26	0.20

2.3 Image processing

The raw images recorded from the experiment cannot be used directly for the bubble shape optimization. An image processing procedure was implemented in MATLAB (R2017b) to extract the bubble profile from the raw images. The specific steps are displayed in **Figure 3**. Firstly, a reference image concerning the background, which was taken before the passage of the bubble, was subtracted from the raw image (**Figure 3-I**). The image thus obtained was then transformed into a binary image (**Figure 3-II**) on which the bubble was depicted by a black spot after the background noise being removed. Since the bubble may slants while rising in the liquid, a rotation was implemented to situate the major axis of the bubble along the vertical direction. The bubble profile (**Figure 3-IV**) was finally extracted by recognizing the edge of the bubble spot (**Figure 3-III**).

Figure 3. Schematic views of the image processing steps (Example of a bubble rising in 0.5% wt. Xanthan solution).

To calculate the velocity of the bubble, the centroid (x_i, y_i) of the bubble spot in each frame of image (**Figure 3-II**) was recognized. Then the distance between the centroids in two successive frames was divided by the time interval Δt (1/2000 s), thus giving the rising velocity of the bubble:

$$U = \frac{\sqrt{(x_{i+1}-x_i)^2 + (y_{i+1}-y_i)^2}}{\Delta t} \quad (2)$$

For a given experimental condition, the averaged values of U were calculated by considering 20 pairs of images, and the standard deviation is smaller than 1.25%. Herein it can be assumed that the velocity on the image area has attained the terminal velocity.

After the image processing, the bubble profile is put in the Cartesian coordinates making the bottom of the bubble coincide with the origin and the major axis of the bubble along the ordinate (see **Figure 4-I**). The coordinates of each point of the bubble profile were then detected to fit the parametric equations which will be discussed in next section.

2.4 Characterization of bubble shapes

2.4.1 Equivalent diameter of the bubble

As shown in **Figure 4-I**, the images obtained from the experiment were two-dimensional bubble profiles. To calculate the equivalent diameter of the bubble, a reconstruction of the three-dimensional bubble was implemented considering that the bubble shape was axisymmetric with respect to the major axis of the bubble profile. Then the 3D bubble was divided into a set of multiple small circular truncated cones (**Figure 4-II**). The volume of each cone equals [36]:

$$V_i = \frac{\pi h}{3} (R^2 + r^2 + R \cdot r) \quad (3)$$

where R and r are the radius of the lower and upper cross-sections, respectively, and h the height of the cone. These three variables could be directly obtained from the bubble profiles (depicted in **Figure 4-I**). The total volume of the bubble is the sum of the all the small cones and the equivalent diameter could be calculated as below:

$$D_{eq} = \sqrt[3]{\frac{6 \sum_{i=1}^N V_i}{\pi}} \quad (4)$$

Figure 4. Schematic diagram of the bubble profile (I) and the processing method for the equivalent diameter of the bubble (II).

2.4.2 Bubble shape equation

As the example shown in **Figure 4**, one of the typical shapes in non-Newtonian elastic fluid is the teardrop-like shape. It would be interesting if this kind of shape could be described by simple parametric equations. On the geometry, there exists several mathematical curves which are similar to the teardrop-like bubble shape, such as teardrop, simple folium and piriform. Their parametric equations are given with corresponding profiles in **Table 2**.

Table 2. Approximate models of the profiles of a bubble or droplet: parametric equation and example.

Teardrop	$\begin{cases} x = a \sin t \left(\sin \frac{t}{2} \right)^n \\ y = a \cos t \end{cases}$	$\begin{aligned} a &= 1 \\ n &= 3 \\ t &\in [-\infty, +\infty] \end{aligned}$
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Simple folium	$\begin{cases} x = \frac{at}{1+t^2} \\ y = \frac{a}{1+t^2} \end{cases}$	$a=1$ $t \in [-\infty, +\infty]$
Piriform	$\begin{cases} x = \frac{a^2}{b} \cos^3 t \sin t \\ y = a \cos^2 t \end{cases}$	$a=1$ $b=1$ $t \in [-\infty, +\infty]$

189

190 It should be noticed that the equations in the table can only be used to characterize the
191 specific shapes displayed in the right-hand side of table (as shown below), i.e. the shapes
192 with cusp at the bottom. In order to extend these equations to be used for more types of
193 shapes like spherical, ellipsoidal and cap bubbles, a modification was implemented on the
194 expression of the parametric equation for folium shape by adding extra variables to the
195 euqation. The appropriate expression of the parametric equation was determined through
196 trial and error. A new parametric model is proposed as below:

$$197 \quad \begin{cases} x(t) = \frac{2x_C t}{1+t^2} \\ y(t) = \frac{y_A}{[1+\alpha t^2]^\beta} \end{cases} \quad t \in [-\infty, +\infty] \quad (5)$$

$$198 \quad E = \frac{2x_C}{y_A} \quad (6)$$

199 where:

- 200 ▪ α, β are two parameters describing the bubble shape;
- 201 ▪ E is the aspect ratio;
- 202 ▪ $2x_C$ is the length of the minor axis of the bubble profile depicted in **Figure 4-I** [m];
- 203 ▪ y_A is the length of the major axis of the bubble profile depicted in **Figure 4-I** [m];
- 204 ▪ t is a numerical variable range between negative and positive infinity.

205 By adjusting α and β , the modified parametric equation can be used to describe the profile of
206 different bubble shapes that encountered in non-Newtonian fluids. The validation of the
207 model is discussed in **Section 3.1**.

208 In a measurement, the lengths of major axis y_A and minor axis $2x_C$ can be obtained directly
209 from the bubble images (**Figure 3-III**). On the other hand, an optimization should be
210 performed to determine the values of α and β . Supposing that the edge of the bubble profile
211 can be divided into N points, the coordinates of point i ($x_{i,exp}, y_{i,exp}$) on the experimental

image are definite. For a pair of arbitrary values (α, β) , from the function of ordinate in **Eq. (5)**, it has:

$$t_i = \sqrt{\frac{1}{\alpha} [(y_{i,exp}/y_A)^{-\frac{1}{\beta}} - 1]} \quad (7)$$

By substituting t_i back to the function of abscissa in **Eq. (5)**, the calculated abscissa $x_{i,cal}$ can be obtained as:

$$x_{i,cal} = \frac{2x_c t_i}{1+t_i^2} \quad (8)$$

The values of the parameters α and β are then optimized by minimizing the sum of the difference at all points between the abscissa calculated with **Eq. (8)** and the one from the experiment:

$$\sum_{i=1}^N |x_{i,cal} - x_{i,exp}| \quad (9)$$

The difference is then compared to the equivalent diameter and the relative deviation of the fitting result is defined as follow:

$$err = \frac{\sum_{i=1}^N |x_{i,cal} - x_{i,exp}|}{N \cdot D_{eq}} \quad (10)$$

3 Results and discussion

3.1 Validation of the parametric model

Firstly, before dealing with the result from the experiment, the proposed parametric model is verified with respect to its capability for characterizing the bubble shape. According to **Eq. (5)** & **(6)**, the parametric equations for the bubble shape involve three parameters: α , β , and the aspect ratio, E . The evolution of the bubble shape in function of α and β is depicted in **Figure 5** in which the profiles of the bubbles are shown for aspect ratio $E = 1$ (**Figure 5-I**) and $E = 0.5$ (**Figure 5-II**). It could be observed that the value E controls the global shape that the profile becomes more oblate with smaller value E (e.g. spherical bubble to ellipsoidal bubble). On the other hand, the parameters α and β control the detail of the shape. The value of α can change the position of the minor axis (horizontal) of the bubble profiles. When the value of β is fixed (e.g. $\beta = 1$) and α increases from 0.5 to 2.5, the minor axis is moving from near the top of the bubble to the bottom of the bubble. On the other hand, β is related to the appearance of the cusp at the bottom of the bubble. Herein, the bubble deformation can be roughly summarized as follows:

- For $\beta < 0.6$, there is a clear cusp at the bottom of the bubble. The typical inverted teardrop shape appears at a high value of α . (e.g. **Figure 5-I**: $(\alpha, \beta) = (2.5, 0.4)$).
- For $0.6 < \beta < 0.8$, the bubble becomes progressively rounded at the bottom and the cusp gradually disappears as the value of β increases.
- For $0.8 < \beta < 1$, there is no longer a cusp and the profile becomes more and more rounded. The quasi-ellipsoidal bubble appears at this interval. In particular, the circular profile (spherical bubble) and the perfectly elliptical profile (ellipsoidal bubble) appear when the value α and β are both equal to 1.
- For $\beta > 1$, the bottom of the profile tends to be flat. The typical cap shape appears when the values of α and β are both sufficient large (e.g. **Figure 5-II**: $\alpha = 1.5, \beta = 1.5$).

Figure 5. Evolution of the shape versus the parameters α and β (I: $E = 1$, II: $E = 0.5$).

Therefore, by adjusting the values of parameters α , β and E , the majority of the bubble shapes appeared in non-Newtonian fluids can be characterized precisely. Based on the parametric equations, the investigation of the bubble shape can be carried out by analyzing the evolution of these parameters.

3.2 Bubble shape regimes

The resulting bubble shapes in different non-Newtonian solutions (i.e. both PAAm and Xanthan gum solutions) are shown as bubble shape regime map, namely so-called Grace Map, in **Figure 6**. This bubble shape regime map is plotted in terms of the dimensionless Re , Eo numbers and $\log Mo$ (dark gray curves), which can be calculated as defined in **Eq.(11)**-

(13), and the typical bubble shapes (gray patterns in the background) extracted from the raw images (Figure 3-III) are also depicted near the corresponding experimental points.

Figure 6. Shape regime map (namely Grace Map) for bubbles in PAAm and Xanthan gum solutions (dark gray curves: $\log Mo$; gray patterns in the background: typical bubble shape near the corresponding experimental points).

$$Eo = \frac{\rho g D_{eq}^2}{\sigma} \quad (11)$$

$$Re = \frac{\rho U D_{eq}}{\mu} \quad (12)$$

$$Mo = \frac{g \mu^4}{\rho \sigma^3} \quad (13)$$

In PAAm solutions, the range of Mo number is relatively small ($-9 < \log Mo < -7$) within the concentration range of study (0.1% - 1.25% wt.). The data covers the spherical bubble and the ellipsoidal bubble regimes. When increasing Eo and Re , the bubble becomes bigger and the shape changes from spherical to ellipsoidal. Regarding the concentration of the solution, it can be seen that the Re increases in a dilute solution, making the bubble shape become more oblate and even causing wobbling due to the surface oscillation.

In Xanthan solutions, the range of Mo number is larger ($-9 < \log Mo < 3$) under the investigated concentration (0.1% - 0.5% wt.). The data covers not only the spherical and ellipsoidal bubbles but also the prolate and teardrop ones. A similar tendency is found for the bubble deformation as in the PAAm solutions. In particular, the cusp appears for high Eo number ($Eo > 1$) and becomes more significant with the increase of the solution concentration or for lower Re number. Under this condition, the bubble is also elongated vertically, which is uncommon in Newtonian fluids.

Compared with the Grace map given by Clift et al. [37] for bubble shapes in Newtonian fluids, the regimes of the spherical bubble and the ellipsoidal bubble in the present study are consistent when $Re > 1$. However, the bubble shapes are invariably spherical when $Re < 1$, which are contrary to those in non-Newtonian fluids. In fact, in non-Newtonian fluids, as the bubble velocity could be extremely low due to the complex rheological properties of the solution, relatively smaller values of Re number should be considered, namely Re ranging from 0.001 to 1 in this study. Within this range, the prolate bubble and teardrop bubble can also be visualized while the spherical bubble appears only for low Eo number ($Eo < 1$).

3.3 Bubble velocity

Concerning the bubble terminal velocity, as shown in **Figure 7**, for both types of the solutions, it first increases with the bubble size then tends to be constant. Although the velocity curves at different concentrations differ more from each other for Xanthan solutions, this evolution and the final constant velocity do not depend on the type of solution or the concentration of the solute. It could be derived that, in a highly viscous Xanthan solution, the bubble rises much more slowly and needs to attain the final constant velocity in a bigger size. In particular, it exists a remarkable difference between the curves of the three lower concentrations of PAAm solutions and those of the two higher concentrations. The reason is that, in the dilute PAAm solutions (0.1%- 0.5% wt), the bubble shape is less stable with the surface oscillation, which leads to the reduction of the viscosity around the bubble due to the shear thinning property. On the other hand, when a bubble is rising in a concentrated PAAm solution (1%- 1.25% wt), the surface oscillation no longer exists due to the higher viscosity. The bubble thus encounters more resistance and its velocity is much slower. Moreover, there is no evident velocity discontinuity under our experimental conditions indicating that the bubble terminal velocity could be less influenced by the bubble shape deformation (from ellipsoidal to inverted teardrop). This result is consistent with the literature [38], in which the same behavior of rising bubbles was found in CMC solutions and Xanthan gum solutions.

Figure 7. Relationship between bubble rising velocity and bubble size ($D_{eq} = 0.7\sim 7$ mm) in PAAm (I) and Xanthan gum solutions (II).

3.4 Bubble shape parameters

To better analyze the bubble shape, the bubble profile was fitted by the parametric models introduced in **Section 2.4.2**. It should be noticed that the bubble shape is assumed to be stable without surface oscillation. The data for big bubble ($D_{eq} > 6$ mm) rising in dilute PAAm solution (0.1%- 0.5% wt) has been eliminated due to the non-negligible surface oscillation. Two typical fitting results, chosen for single bubbles in Xanthan gum and PAAm solutions, are shown in **Figure 8-I & II**, where the black dots depict the origin profile of the bubble and the red line is the fitting profile with the values of α and β given alongside. It can be seen that whether the bubble has a teardrop or an ellipsoidal shape, the deviation between the experimental points and the fitting points is small. The deviation defined in **Eq. (10)** is in a magnitude smaller than 3% for all the cases. To ensure the validity of the bubble characterization result, fitting data with an error of more than 2% was eliminated. An example of the averaged fitting results of the bubble shapes in both PAAm and Xanthan solutions under different concentrations are shown in **Figure 8-III & IV**. It can be observed that under the same operational condition for bubble generation (i.e. the size of nozzle and the air flow rate), the sizes and shapes of bubbles depend on the type and concentration of the solutes (i.e. PAAm and Xanthan gum). In addition, the evolution of the bubble shapes given by the model is consistent with the original bubble shapes depicted in **Figure 6**.

Figure 8. Examples of the bubble profiles and the fitting ones by Eq. (5) & (6) in PAAm (I, III) and Xanthan solutions (II, IV) under different concentrations. (The bubbles are injected by the nozzle with a caliber of 1 mm at a flow rate of 10 $\mu\text{L/h}$).

It can be seen from **Figure 8-III & IV** that in PAAm solution, the size of the bubble slightly changes, while the shape becomes more and more rounded for higher concentrations, namely, from an oblate bubble to a general ellipsoidal bubble. In contrast, in Xanthan solutions, the major axis (vertical) of the bubble increases as the concentration of Xanthan increases, as well as the bubble deforms from an oblate ellipsoid to an oblate teardrop and then turns to an alongate teardrop.

For a single bubble, a pair of parameters (α , β) could be obtained from the fitting result. In **Figure 9**, the relationship between the bubble shape parameters (α and β) are depicted for bubbles in both PAAm and Xanthan gum solutions. The corresponding bubble profiles (light gray outline) are also displayed at the positions of values α and β . In this figure, the delicate changes of bubble shape can be visualized and quantified, which is difficult to achieve simply by using the aspect ratio. For PAAm solutions, the data are concentrated in the range of $1 < \alpha < 1.2$ and $0.8 < \beta < 1$, suggesting that the bubble is spherical or ellipsoidal, while, for the Xanthan solution, the range of these two parameters is much larger. As the parameter α increases, the value of β decreases gradually. As interpreted in **Section 2.4.2**, the cusp of

the bubble appears when $\beta < 0.6$ and it becomes sharper when β continues to decrease. The result of the Xanthan gum cases shows good agreement with this evolution.

Figure 9. Relationship between the bubble shape parameters (α and β) in both PAAm and Xanthan solutions under different concentrations.

It is found that the relationship between α and β could be correlated by a power-law function, which is expressed as below:

$$\alpha^{0.85}\beta = 1 \quad (14)$$

In the range investigated in this study ($-9 < \log Mo < 3$), the accuracy of this correlation is acceptable, with a determination coefficient $r^2 > 95\%$. It can be seen from **Figure 9** that the curve of **Eq. (14)** passes the point $(\alpha, \beta) = (1, 1)$, which indicates a perfect spherical bubble (or an ellipsoidal bubble when $E \neq 1$). Thanks to **Eq. (14)**, it is possible to characterize the bubble shape with only one parameter (either α or β). In the next section, only the parameter β is taken into account.

3.5 Prediction of bubble shape

It is known from **Section 3.1** that the aspect ratio E roughly describes the bubble shape while the parameter α and β characterize the details of bubble deformation. As mentioned in the introduction that a lot of research has been carried out for predicting the aspect ratio E for bubbles in different liquids, we will focus on the discussion of the bubble shape parameter β .

Depending on the value of β , bubble shape changes from ellipsoidal (without cusp) to teardrop (with cusp). Some existed research attempts to explain the appearance of the cusp

theoretically and most of the authors support the reason that the majority of non-Newtonian fluids are composed of long-chain molecules and possess viscoelasticity [39,40]. To quantify the elasticity of the fluid, the Deborah number is utilized to compare the relaxation time of the fluid to the time scale of observation [41]:

$$De = \frac{\lambda U}{D_{eq}} = \lambda \dot{\gamma} \quad (15)$$

In our study, the observation time is estimated that it is equal to the characteristic time of the motion (i.e. the equivalent diameter of the bubble divided by the bubble rising velocity). Since this characteristic time of the motion is relatively short comparing with the relaxation time of the fluid, the magnitude of calculated De number in this study may be higher than the literature [42]. Nonetheless, it can still be used to compare the elastic levels for the investigated fluids.

Since the cusp could now be quantified by the parameter β , the evolution of β is analyzed in terms of the bubble size (V) and various dimensionless numbers (De , Eo , Re), shown as in **Figure 10**.

Figure 10. The evolution of parameter β in terms of the bubble size (V) and various dimensionless numbers (De , Eo , Re) in PAAm and Xanthan solutions under different concentrations.

It can be obtained from **Figure 10** that: (1) for larger bubbles, the velocity of the bubble increases then the bubble begins to deform and becomes less rounded when the contribution of inertial force and buoyancy is stronger than the surface tension and viscous force. (2) β decreases with higher elasticity of the solution in most investigated range except for some PAAm which are relatively less elastic. If the bubble moves too fast, the liquid cannot fill the space in the rear of the bubble immediately. Hence this space is still occupied by the gas and pressed by the long-chain liquid molecule from the side, leading to a little cusp forming in the rear of the bubble. (3) In the evolution of $\beta - De$ and $\beta - Re$, the evolution of the value β under three concentrations is quite dispersed. This result might be caused by the different shear-thinning level of the solutions as the shear-thinning property is also a reason for the deformation of the bubble [43].

The data in **Figure 10** are too scattered for a general law to be obtained. Hence multiple influencing factors are considered instead of one. By analogy with the analysis of aspect ratio, the investigation was carried out for the evolution of β with different pairs of dimensionless numbers as well as the shear-thinning property which can be roughly quantified by the power index n (Eq. (1)). After testing several functions, the following correlation was proposed to fit the experimental data:

$$\beta = n - 0.11De^{0.19}Eo^{0.11} + 0.80 \quad (16)$$

This equation is correlated to the result depicted in **Figure 10** as β decreases monotonically with high De , Eo numbers and the shear-thinning levels. The comparison between parameter β predicted by **Eq. (16)** and the experimental ones is shown in **Figure 11**. It can be seen that, under the range of this study ($-9 < \log Mo < 3$), the prediction quality of parameter β in both PAAm and Xanthan gum solutions is satisfactory. Relatively large deviation appears at high values of β due to the wobbling of the bubble rising in dilute PAAm solutions. The fitting result has a coefficient of determination (r^2) higher than 95%.

Figure 11. Comparison result between parameter β from correlation (Eq.(16)) and β from measurement in PAAM and Xanthan gum solutions.

4 Conclusions

In this paper, various single air bubbles rising in stagnant non-Newtonian fluids (polyacrylamide, Xanthan gum) were investigated by high-speed photography. The shapes of the bubbles from spherical to inverted teardrop can be characterized with good accuracy by parametric equations which contain the aspect ratio (E) and two shape parameters (α , β). Based on this method, the various impacts on the bubble shape can be quantified by studying the evolution of the three parameters as a function of bubble size, bubble velocity, multiple dimensionless numbers (Re , Eo , etc.) and the property of the fluids. Within the range investigated here ($-9 < \log Mo < -7$), it is found that the cusp of the bubble (quantified by β) is related to the viscoelasticity as well as the shear-thinning level of the solutions. A simple correlation is then proposed to predict the parameter β . Since the parametric equations are capable to characterize most of observed shapes (including cap bubble), this method can be applied to bigger bubble or droplets in complex Newtonian and non-Newtonian fluids.

442 Symbols used

D_{eq}	[mm]	equivalent diameter
E	[-]	aspect ratio
Eo	[-]	Eötvös number
g	[m·s ⁻²]	gravitational acceleration
Mo	[-]	Morton number
n	[-]	power index / shear-thinning level
P	[mm]	perimeter
r^2	[-]	determination coefficient
Re	[-]	Reynolds number
S	[mm ²]	surface area
t	[-]	numerical variable
Ta	[-]	Tadaki number
U_x	[mm·s ⁻¹]	oscillating velocity
U_y	[mm·s ⁻¹]	rising velocity
V	[mm ³]	Bubble volume
We	[-]	Weber number
$x_{i,cal}$	[mm]	abscissa by calculation
$x_{i,exp}$	[mm]	abscissa by experiment
x_C	[mm]	half-length of major axis
y_A	[mm]	length of minor axis

Greek Symbols

α	[-]	bubble shape parameter
β	[-]	bubble shape parameter
$\dot{\gamma}$	[s ⁻¹]	shear rate
λ	[s]	relaxation time
μ	[Pa·s]	dynamic viscosity
ρ	[kg·m ⁻³]	density
σ	[N·m ⁻¹]	surface tension

443

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