Bubble formation on submerged micrometer-sized nozzles in polymer solutions: An experimental investigation

Amirmohammad Sattari, Pedram Hanafizadeh

School of Mechanical Engineering, College of Engineering, University of Tehran, Iran

GRAPHICAL ABSTRACT

The contribution of effective forces acting on the bubble near detachment moment on 450 mm nozzle and 0.5 ml/min gas flow rate.

ARTICLE INFO

Keywords:
Bubble formation
Micro-Nozzle
Non-Newtonian fluids
Young-Laplace equation
Necking phenomenon

ABSTRACT

This research presents detailed formation of air bubbles on different submerged micrometer-sized nozzles in Newtonian and non-Newtonian fluids. An experimental study is performed using the high-speed camera on three nozzles with diameters of 150, 450, and 600 μm under relatively low gas flow rate conditions (0.1 – 1 ml/min). Distilled water and various concentrations of Carboxymethyl cellulose (CMC) aqueous solutions are utilized as the continuous phase for each type of fluid. Different characteristics of the bubble formation such as volume change, frequency, height, maximum width, and instantaneous contact angle are obtained using an image processing technique and the Young-Laplace equation. Effect of different parameters including gas flow rate, nozzle diameter, concentration, and rheological properties of the continuous phases are investigated. Also, a detailed dynamic force analysis is performed to survey the role of fluid properties on the effective forces. The force balance reveals that the surface tension force is dominant in the smallest orifice, while its influence decreases as the orifice diameter increases. Moreover, it was observed that the drag and hydrostatic forces have relatively noticeable contributions in this low gas flow rates. Furthermore, results show that under the conditions of this research, the characteristics of the bubble are weakly dependent on the gas flow rate. On the other hand, the concentration of the CMC and the rheological behavior of the non-Newtonian fluids so-obtained have a significant influence on bubble formation process. Also, observations made and predictions done as to the bubble's curvature (say, based on Young-Laplace equation) suggest that the necking phenomenon does not exhibit itself at the top of the nozzle in non-Newtonian liquids flowing through micron-sized nozzles.

https://doi.org/10.1016/j.colsurfa.2018.12.029

Received 1 October 2018; Received in revised form 8 December 2018; Accepted 13 December 2018

Available online 14 December 2018

© 2018 Elsevier B.V. All rights reserved.
1. Introduction

Bubbly flow is one of the major flow patterns of two-phase flow which plays a significant role in various applications concerning with dispersion of gas bubbles in liquids. The dynamic of the single bubble is one of the most important section of this flow pattern. This phenomenon is relatively complicated and affected by many parameters such as orifice diameter, gas flow rate, liquid height, wettability, and physical properties of the dispersed and continuous phases.

Bubble departure volume (volume of the bubble at the detachment moment) and the influence of different parameters on it are one of the main issues in bubble formation topic. There is an obvious difference between the trend of bubble volume departure on millimeter and micrometer-sized orifices. The bubble volume was reported to increase with increasing gas flow rate in millimeter-sized nozzles in many studies [1–4] and in our previous works [5,6]. This is in contrast with previous studies that were conducted on micrometer-sized nozzles, in which it was reported that volume of the bubble follows a U-shaped trend with increasing gas flow rate [7,8].

Although there are many works done in the field of bubble formation especially capillary tubes, most of the works focus on millimeter-sized nozzles. In the case of micrometer-sized nozzles, Datta et al. [7] were the first one who observed the U-shape behavior of the bubble departure volume versus gas flow rate in a micronozzle. A similar observation has been made by Vafaei and Wen [8]. They investigated the formation of a bubble in micronozzle with a radius of 55 μm in distilled water. They utilized the Young-Laplace equation to predict the bubble evolution and curvature and found that it can predict that quite well until the detachment moment. Also, they observed that bubble volume obeys a U-shaped trend with increasing of gas flow rate, which is in contrast with previous studies done in millimeter-sized nozzles. In another research done by Vafaei et al. [9] in a substrate micrometer-sized nozzle, the formation of the bubble has been investigated experimentally. They utilized nozzles with an internal diameter sized in the range of 0.11–0.84 mm and relatively low gas flow rates (0.015 to 0.85 ml/min). They observed that the bubble expansion rate follows cyclic growth and decay periods until the bubble finally detaches for the substrate nozzle. They found that these fluctuations are related to the rapid increase of the inertial force during the early stage of bubble formation.

The material of the nozzle has a considerable effect on bubble formation. Lin et al. [10] investigated the influence of contact angle on bubble formation. They found that the surface wettability had the most important role in bubble size. Davidson and Amick [11] observed that at relatively low gas flow rates, bubble volume is a function of surface tension and orifice diameter, and is independent of gas flow rate. Furthermore, as the gas flow rate increases, the bubble volume becomes independent of surface tension and only is a function of gas flow rate. Corchero et al. [12] studied bubble formation on orifices with different materials with contact angles of 68° ≤ θ ≤ 123°. It was shown that data for different contact angles and orifice diameters can be approximately reduced to a single relationship between bubble volume and flow rate when a properly-scaled bubble volume at detachment was plotted versus a properly scaled volumetric gas flow rate. In a series of studies, Gnyoskureno et al. [13–15] observed that wettability has considerable influence on bubble volume departure; the bubble volume increased by roughly 50% as contact angle increased from 68° to 110°.

Bubble formation in non-Newtonian fluids has attracted many attentions due mainly to its applications. Ellis and Waugh [16] were the first who studied bubble cavitation in non-Newtonian fluids. In another research, Ting and Ellis [17] conducted an experimental investigation on bubble cavitation in polymer solutions. They observed a retardation in the growth rate of the bubbles in polymer solutions in comparison with water. The effect of viscoelasticity on the non-spherical bubbles dynamics was studied by Hara and Schowalter [18]. They showed that rheological properties of a non-Newtonian fluid have a significant affection, especially on non-spherical bubbles. Terasaka and Tsuge [19] investigated the effect of orifice diameter on the bubble dynamic in non-Newtonian fluids. They found that at relatively low gas flow rates, bubble volume increases with decreasing orifice diameter while in high gas flow rates, orifice diameter does not have a noticeable impression on bubble volume. However, Badam et al. [20] observed that in Newtonian fluids, at any flow rate, bubble departure volume increases with increasing gas flow rate. Another study on bubble formation in the non-Newtonian fluid was performed by Fan et al. [21]. They perceived that bubble departure volume increases with increasing concentration of the solution and orifice diameter.

Most of the studies in the field of bubble formation have been conducted on millimeter-sized nozzles, and there are a few studies investigated this phenomenon on micrometer-sized nozzles, especially in non-Newtonian fluids. There is a gap in knowledge related to the bubble formation on micrometer-sized nozzles in non-Newtonian fluids, at least, to the best knowledge of the authors. With the aim of filling this gap, in the present work, the formation of bubbles on micrometer-sized nozzles submerged in non-Newtonian fluids has been studied experimentally at relatively low gas flow rates. The effects of different parameters such as nozzle diameter, gas flow rate, concentration, and rheology properties of the continuous phase have been investigated on major characteristics of a bubble during its evolution and especially near detachment time. Image processing technique and the Young-Laplace method are utilized for extracting the characteristics of the bubble. Besides, a detailed dynamic force analysis is done in order to examine the influence of the continuous phase rheology on the forces acting effectively on the bubble.

2. Methodology

2.1. Experimental setup

Fig. 1 illustrates a schematic of the experimental apparatus. The experimental setup includes a square column, micronozzle, an automatic syringe pump for gas flow injection, a high-speed camera, and a LED lamp. The nozzles are made of polymethyl methacrylate (PMMA) with a precision laser cutting method and have three different internal diameters, namely 150, 450, and 600 μm. Micronozzle is submerged into a square column of fluid made of PMMA with dimensions of...
20 mm × 20 mm × 100 mm which is open to the atmosphere at the top. The square column is filled with quiescent distilled water or non-Newtonian fluids to a height of 50 mm above the tip of the nozzle. The air flow is supplied from an automatic syringe pump in the range of 0.1−1 ml/min. A high-speed camera with 1200 fps speed and a LED lamp are utilized for capturing and recording the details of the bubble formation mechanism. The images are stored on a PC and are analyzed by using an image processing code. Also, all experiments were conducted three times in order to make sufficient reproducibility and the reported data are an average of the results of different tests.

Fig. 2a shows the dimensions and schematic of the nozzles. At first, the working fluid enters in a cylinder with a diameter of 1 mm, then from a sudden contraction route is forced to flow into the micrometer-sized cylinder with 150, 450, and 600 μm internal diameters. The nozzles were made of PMMA material with CO$_2$ laser cutting. A contact angle meter is used to evaluate the wettability of the PMMA material. The droplet contact angle of 8 μl volume on the PMMA material is observed to be around 72° (Fig. 2b), so that the nozzle can be categorized as a hydrophilic surface.

The Carboxymethyl cellulose (CMC) aqueous solutions are used as the non-Newtonian continuous phases with various concentrations in distilled water (0.1%, 0.25%, and 0.5% wt). These solutions have high viscosity and low elasticity. The rheological properties of the non-Newtonian fluids are shown in Fig. 3, which were gathered using a rheometer. As illustrated, the CMC solutions show shear thinning behavior. As is well-established in the literature, CMC solutions are non-Newtonian and obey the Carreau model [22], which reads as:

\[ \frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} = (1 + (\lambda \dot{\gamma})^2)^{(n-1)/2} \]

where $\eta$ is the viscosity of the fluid, $\eta_\infty$ is the viscosity of the solution at an infinite shear rate, which is usually considered to be the viscosity of the solvent (distilled water) [23], and $\eta_0$ is the viscosity of the solution when shear rate approaches zero. By fitting appropriate curves to the data of Fig. 3, the two parameters of this model ($\lambda$ and $n$) are obtained, which are tabulated together with other properties of the solutions in Table 1. As it can be seen in this table, by incremental of the CMC concentration, the fluids’ viscosity and interfacial tension are both increased, however, the interfacial tension is a weak function of concentration.

2.2. Force acting on a growing bubble

Characteristics of the bubble during its evolution are related to the contribution of forces acting on it. These forces include the buoyancy force due to the difference of density between continuous and dispersed phases, the interfacial tension force, due to the Young–Laplace pressure, the hydrostatic force of the gas and liquid phases’ momentum, and the
Viscous drag force created by the continuous phase. The effective forces on the bubble are depicted in Fig. 5 and are formulated in below.

The buoyancy force is defined as,

$$F_g = \rho_l \cdot g \cdot V$$  \hspace{1cm} (2)

where $V$ is the volume of the bubble.

The force due to the Young-Laplace pressure is generally expressed as,

$$F_p = \frac{2\sigma \cdot R_1 \cdot R_2}{R_{eq}}$$  \hspace{1cm} (3)

in which $\sigma$ and $r_e$ are interfacial tension and radius of the contact line, respectively. $R_1$ and $R_2$ are radii of curvatures which describe the latitude and longitude of the curvature as it rotates, respectively [8].

For calculating radii of curvatures, as illustrated in Fig. 4, the bubble curvature is divided into two-dimensional axisymmetric elements. $R_2$ is the radius of the circle which passes through the elements $(j-1)$, $j$, and $(j+1)$, and has a center point of $O$; and $R_1$ is the distance from the bubble axis of symmetry to the element $j$ through the point $O$. The average of all equivalent radius $R_{eq}$, which is defined by the following equation is used as the characteristic radius for the term of different Laplace pressure [6]:

$$R_{eq} = \frac{1}{2} \left( \frac{1}{R_1} + \frac{1}{R_2} \right)$$  \hspace{1cm} (4)

Therefore, the pressure force can be written in the following format:

$$F_p = \frac{2\sigma \cdot R_1 \cdot R_2}{R_{eq}}$$  \hspace{1cm} (5)

When a gravity field presents, the hydrostatic force acting on the bubble appears in the direction of the gravitational field, which causes the bubble shape to deviate from its spherical shape. The hydrostatic force can be expressed using the following equation [4]:

$$F_h = (\rho_l - \rho_g) \cdot g \cdot h \cdot \pi \cdot R^2$$  \hspace{1cm} (6)

in which $h$ is the height of the bubble.

The vertical component of the surface tension force is related to the contact perimeter of the bubble and contact angle,

$$F_c = 2\sigma \cdot \pi \cdot \sin \theta$$  \hspace{1cm} (7)

in which $\theta$ is the contact angle of the bubble.

The kinetic or momentum force ($F_M$), resulting from the fluid motion in the capillary, which tends to separate the bubble from the nozzle, can be calculated using the following equation [24]:

$$F_M = \rho_g \cdot U_g \cdot Q$$  \hspace{1cm} (8)

where $Q$ is the nominal volumetric flow rate of the gas phase and $U_g$ is the superficial velocity of the gas phase ($U_g = 4Q/\pi D^2$), in which $D$ is the inner diameter of the nozzle.

The drag force in non-Newtonian fluids is one of the concerning issues due to the variable viscosity of the fluid. There are some works which present models that can predict the drag force in the non-Newtonian fluids based on the Carreau model. Rodrigue et al. [25] presented a model for obtaining drag force in non-Newtonian fluids as a function of the shear-thinning character:

$$F_D = 4\pi \cdot \eta_0 \cdot R_e \cdot \left[ 1 + \frac{4}{25} \left( \frac{\mu}{\pi} - 1 \right) \cdot Cu \right]$$  \hspace{1cm} (9)

where

$$Cu = \frac{\lambda \cdot U_b}{R_e}$$  \hspace{1cm} (10)

$Cu$ is Carreau number which is a representation of variable viscosity, $U_b$ is the bubble velocity, $R_e$ is the equivalent bubble radius, $\lambda$ and $\eta$ are parameters of the Carreau model, and $\eta_0$ is the viscosity of the solution when shear rate approaches zero.

For the Newtonian fluid, the drag force is obtained utilizing the following equation:

$$F_D = \frac{1}{2} \cdot \rho_l \cdot \pi \cdot W_{max} \cdot U_b^2 \cdot C_D$$  \hspace{1cm} (11)

in which $W_{max}$ is the maximum width of the bubble, which is considered due to this fact that the drag force acts on the projection area of
the body. Also, \( C_D \) is the drag coefficient that can be calculated using Sekoguchi correlation [26] for \( 1 < Re_b < 1000 \):

\[
C_D = \frac{18.5}{Re_b}
\]  

where \( Re_b \) is the Reynolds number of the bubble in Newtonian fluid \((Re_b = \dot{V} d_b / \eta)\), and \( d_b \) is the equivalent diameter of the bubble.

Therefore, an equilibrium force balance takes the below form,

\[
\sum F = F_S + F_F + F_M - F_C - F_c - F_B = 0
\]  

(13)

2.3. Analysis of axisymmetric bubble shape

In order to predict the shape of the axisymmetric bubble, the Young-Laplace equation is employed. By considering the start of the axis at the bubble apex (see Fig. 5), the Young-Laplace equation can be written as the following format [9]:

\[
\frac{d\theta}{ds} = \frac{2}{R_0} - \frac{g z}{\sigma} (\rho_l - \rho_g) - \frac{\sin \theta}{r}
\]  

(14)

where \( R_0 \), \( \theta \), and \( s \) are the radius of the curvature at the bubble apex, contact angle, and curve length at the point \((r, \theta)\). The coordinate of an axisymmetric bubble is depicted in Fig. 5.

The Eq. (14) can be solved along with the following system of differential equations in order to obtain the bubble shape up to the detachment moment [9]:

\[
\frac{dr}{ds} = \cos \theta
\]  

(15)

\[
\frac{dz}{ds} = \sin \theta
\]  

(16)

\[
\frac{dV}{ds} = \pi r^3 \sin \theta
\]  

(17)

This system of ODEs avoids the singularity problem at the bubble apex, because [8]

\[
\frac{\sin \theta}{r} = 0 = \frac{1}{R_0}
\]  

(18)

By knowing two parameters of the bubble characteristics (such as the radius of contact line, contact angle at the triple line, bubble volume, or location of the apex) at each moment during the bubble growth, the system of ODE of Eqs. (15)-(17) can be solved to obtain the bubble shape, utilizing the following boundary conditions [8,9,27]:

\[
r(0) = z(0) = \theta(0) = V(0) = 0
\]  

(19)

The accuracy details of the Young-Laplace equation in Newtonian fluids can be found in [8,28]. Here we consider bubble height and its maximum radius, which can be exactly measured from the experiments, as two input parameters of the equations and therefore the rest of parameters are extracted by solving Young-Laplace equation in each time step. The set of ODEs are solved in Matlab software using ODE45 integration routine. Also, by comparing the results obtained by the Young-Laplace equation and experimental observation, the accuracy of the model in non-Newtonian fluids will be evaluated.

3. Results and discussion

3.1. Bubble characteristics

Generally-speaking, the formation of a bubble can be divided into three stages: waiting, expansion, and detachment. Fig. 6 shows the sequences of bubble formation from the beginning of the expansion stage up to the detachment moment in distilled water and non-Newtonian fluids with various concentrations on 450 μm nozzle and 0.5 ml/min nominal flow rate as a sample test case. Real and non-dimensional times are shown above each sequence. Non-dimensional time is defined as \( t* = t/\tau \), in which \( \tau \) is the total duration of bubble formation. As it can be seen, the largest duration of the bubble formation is assigned to the waiting stage (the time period without bubble growth). This is in contrast with previous observations in millimeter-sized nozzles which mentioned that the expansion stage has the longest duration among other stages [4,29,30]. This can be justified noting the fact that the surface forces are dominant in formation of the bubble on the micrometer-sized nozzle, which result in a long time of bubble retention time. Based on this figure, an increase in the viscosity of the continuous phase proceed to a larger portion of the formation time in the expansion and detachment stages. It means that for the same nozzle diameter and gas flow rate, generally, departure volume increases with increasing viscosity. This can be attributed to the incremental of viscous drag and the interfacial tension forces (see Table 1) which are forcing the bubble to stick more to the nozzle. This leads to the higher buoyancy force which overcomes downward forces and consequently larger bubble departure volume.

For evaluation of the accuracy of the Young-Laplace equation, especially in a non-Newtonian fluid, the evolution of bubble formation predicted by Young-Laplace equation in water and 0.5% wt CMC solution up to the detachment moment is plotted and compared with experimental observation in a gas flow rate of 0.5 ml/min and 450 μm nozzle diameter in Fig. 7. It is obvious that the theoretical predictions of the Young–Laplace equation shows a good agreement with the experimental results in both Newtonian and non-Newtonian fluids. It was predictable because there is no assumption on the Newtonian behavior of the fluid in driving bubble shape from the Young-Laplace equation.

Fig. 8 illustrates the departure volume of the bubble as a function of the nominal flow rate and nozzle diameter in distilled water. The bubble detached volume generally decreases at low flow rates, then reaches a minimum value which depends on the nozzle diameter, and then immediately starts to increase. This behavior can be explained by considering the dual effects of the flow rate. Firstly, increasing the flow rate result in increasing the amount of the injected flow into the bubble at a specific time. Secondly, increasing the flow rate has an influence on expansion and detachment processes due to increasing pressure and momentum forces, which intend to reduce expansion time and consequently the departure volume as well. In practice, the bubble departure volume is affected by a combination of these two effects. On the other hand, the variations of bubble departure volume from its mean value are ~21%, ~17%, and ~6% in 150, 450, and 600 μm nozzles, respectively. This shows a relatively low dependency of the volume on the flow rate. However, as the nozzle diameter decreases, the influence of the flow rate on the departure volume increases. It is clear that with increasing nozzle diameter, generally, bubble departure volume increases due to an increase of the perimeter which consequently result in larger surface tension force. But, as shown in Fig. 8, there is a critical flow rate (~0.4 ml/min) at which an abnormal behavior of the bubble departure volume can be seen. At this condition the departure volumes of the bubbles that are formed on 600 μm nozzle are lower than ones generated on 450 μm nozzle before this flow rate.

For a better understanding of this behavior Fig. 9 has been plotted, which shows the variation of bubble volume with the formation time (Fig. 9a) and the normalized volume with the normalized time (Fig. 9b) in different nozzles. It is clear that the slope of this figure shows the growth rate of the bubble volume and therefore, as the nozzle diameter decreases, formation time increases. As illustrated in this figure, although the formation times of the bubbles formed on 150 μm nozzle are longer than those for 450 μm and 600 μm nozzles, with the smaller ultimate volumes. This can be justified with this fact that the less portion of the formation time belongs to the expansion stage in smaller micronozzle and most of the formation times belong to the waiting stage, in which no bubble evolution occurs. Generally, two major parameters should be considered simultaneously for evaluating bubble volume. The first one is the total expansion time at detachment stages,
and the second one is the expansion rate. Shorter expansion time and higher expansion rate may result in smaller or larger bubbles. Although more portions of the bubble formation stages are belonging to the expansion and detachment phases for the bubbles formed on the 600 μm nozzle than the 450 μm nozzle (see Fig. 9b), the higher expansion rate of the bubbles formed on 450 μm nozzle leadsto the higher detached volume in this nozzle at nominal flow rate of 0.2 ml/min. It should be noted that if the beginning of the time is considered to be at the beginning of the formation stage (neglecting the waiting time), at the same time, the bubble volume is larger in 600 μm nozzle than the 450 μm nozzle because the larger rim of the bubble is due to the larger perimeter of the bigger nozzle. While the higher expansion rate in smaller nozzle resulted in lower ultimate volume in the larger nozzle.

It should be noted that when a bubble forms at the top of the nozzle, the nominal and the average gas flow rates are slightly different. The average gas flow rate is calculated from $Q_{av} = V_b f$, in which $V_b$ is the bubble volume near detachment moment and $f$ is bubble frequency [9]. For instance, for 450 μm nozzle diameter and 0.2 ml/min nominal flow rate, the bubble volume near detachment moment in distilled water is measured to be 11.0705 mm³ (Fig. 9) and frequency of formation is

---

Fig. 6. Sequences of bubble formation in: (a) distilled water, (b) 0.1% wt CMC, (c) 0.25% wt CMC, and (d) 0.5% wt CMC on 450 μm nozzle and 0.5 ml/min nominal flow rate.
Based on the mentioned values, the average flow rate will be 0.18 ml/min, while the nominal flow rate is 0.2 ml/min. This difference arises from the function of a syringe pump, i.e., using a syringe pump does not assure a stable flow rate or a stable pressure because the gas is compressible and the air pressure at the exit of the nozzle is unstable due to the bubble formation process. So, the syringe pump only assures a time-averaged flow rate. This behavior leads to the non-linear changes of bubble volume versus the formation time. It should be noted that the flow rates which are mentioned in this paper are the nominal flow rates.

A similar behavior is observed in non-Newtonian fluids in Fig. 10 for different concentrations. The comparison between Figs. 8 and 10 shows that the critical flow rate is increased from Newtonian fluid to non-Newtonian ones. Moreover, the critical flow rate is increased with increasing of CMC concentration. The increase in viscosity in these solutions and the lower strain rates [31] may be the reason for such a retardation in the critical flow rate. Also, with increasing viscosity of the continuous phase, the mentioned critical flow rate increases (from about 0.6 ml/min in 0.1% wt CMC solution to the 0.8 ml/min in 0.5% wt CMC solution). On the other hand, with increasing viscosity, bubble departure volume of the 150 μm nozzle gets closer to the bigger nozzles, while in 0.5% CMC concentration, the detached bubble volume of 150 μm nozzle is higher than the other ones. It can be concluded that with increasing viscosity and interfacial tension of the continuous phase (see Table 1), viscous drag and interfacial tension forces are more dominate than the buoyancy and pressure forces during the bubble evolution in the smaller nozzle and result in larger buoyancy force which overcome downward forces and consequently bigger departure volume. Fig. 11 confirms the mentioned claim and shows the volume of the bubble versus time (Fig. 11a) and normalized volume versus normalized time (Fig. 11b) for 0.5% wt CMC solution, 0.5 ml/min flow rate, and different nozzle diameters. As can be seen, formation time increases with decreasing nozzle diameter similar to the distilled water. In addition, the expansion rate increases dramatically from 600 μm to 150 μm nozzles, while expansion time decreases. Simultaneous influences of expansion rate and expansion time resulted in larger bubbles on 150 μm nozzle in this concentration.

Fig. 12 illustrates the variation of the detached volume versus gas flow rate in different continuous phases for 450 μm nozzle. It is clear that with increasing concentration of the continuous phase, and consequently viscosity and interfacial tension, the detached volume increases due to the incremental of the downward forces. As the flow rate increases, the detached volumes at different continuous phases get closer to each other. This is due to the shear-thinning behavior of the non-Newtonian fluids which was shown in Fig. 3. As the flow rate increases, velocity and consequently shear rate between phases increase and resulted in lower viscosity, however, the value of the interfacial tension is not significantly affected with the CMC concentration. Therefore, the influence of the viscosity on departure volume decreases with increasing flow rate in shear-thinning liquids.

Fig. 13 shows the variation of the Weber number ($We = \rho_U l^2 dB / 2\sigma$) with the Reynolds number ($Re = \rho l U g / \eta$) in different continuous phases, where $\rho$, $U$, and $dB$, represent continuous phase density, superficial velocity, and bubble diameter just before detachment moment respectively. Note that the Reynolds number for shear-thinning fluids is

![Fig. 7. Comparison between experimental bubble shapes and bubble evolution predicted by Young–Laplace equations for (a) water and (b) 0.5% wt CMC at a nominal gas flow rate of 0.5 ml/min and 450 μm nozzle diameter.](image1)

![Fig. 8. Detached volume versus flow rate and nozzle diameters in distilled water as the continuous phase.](image2)
defined using the following equation [32]:

\[ Re = \frac{\rho_g U^2 d_n^4}{k} \]  \hspace{1cm} (20)

where \( \rho_g \) is the density of the gas phase. The constants \( k \) and \( n \) were determined from the fits of the Carreau model to the fluid viscosity-shear rate curve as described before, with \( k = \eta_0 \lambda n^{-1} \) being the fluid consistency.

As can be seen in Fig. 13, We number increases with increasing Re number. As the Re number increases, inertial forces are more dominate than viscous ones. Also, increasing We number resulted in the domination of inertial forces to the surface tension ones. It shows that with
increasing Re and We numbers, contributions of surface tension and viscous forces reduce in comparison with inertial ones, which generally leads to the lower detached volume. On the other hand, in non-Newtonian fluids and in a certain Re number, a higher concentration of the continuous phase cause higher We number at detachment moment. For instance, at a constant Re number of approximately 50, the We numbers for 0.1%, 0.25%, and 0.5% wt of CMC solutions are approximately 0.3, 2.5, and 27 respectively. In contrast, again in non-Newtonian fluids, when We number is considered to be constant, Re number decreases with increasing of the concentration. For example, at a constant We number of approximately 5, the Re numbers for 0.1%, 0.25%, and 0.5% wt of CMC solutions are approximately 1000, 88, and 20 respectively. Besides, it can be inferred from the figure that the trend of changing procedure of the Re number with We number is similar in both Newtonian and non-Newtonian fluids.

The frequency of bubble formation is another major characteristic of the bubble which is shown in Fig. 14 versus flow rates in different liquids on 450 μm nozzle. This figure reveals that the frequency of bubble formation increases continuously with increasing gas flow rate and decreases with increasing concentration of the continuous phase. It was predictable because the formation frequency and bubble volume have an inverse relation in these low flow rates. The change in the frequency of bubble formation appears to be quite linear in distilled water for these relatively low flow rates (<1.0 ml/min). This has been confirmed before with various studies [11,33]. However, fitting second-order polynomial equations show the better coefficient of determination ($R^2$) than the linear one in non-Newtonian fluids. So, it can be concluded that the bubble formation frequency in non-Newtonian fluids, unlike Newtonian ones, has a nonlinear dependence on gas flow rate. This may have been caused by the variable viscosity in different flow rates that is because of the consequence of the shear rate changes.

Fig. 15 shows the variation of the instantaneous contact angle as a function of normalized time at different continuous phases. Similar to the previous studies, contact angle exhibits a U-shaped dependency on time. As illustrated, bubbles formed in water (Newtonian fluid) have an obtuse angle near detachment time due to the necking process. Very close to the detachment moment, the bubble is in the process of detaching by forming a neck near the bubble base, in which the influence of buoyancy is enhanced as the curving inward of the neck increases the volume being acted upon by buoyancy. This causes the upper region to
accelerate upward. However, it is interesting to note that bubbles which are generated in non-Newtonian fluids have an acute angle at the detachment moment. This conclusion is based on both observation and applying the Young-Laplace equation (Fig. 7). This fact shows that the necking process does not happen on the top of the nozzle surface in these fluids since the necking process happens when the instantaneous contact angle changes from an acute angle to the obtuse one. It is suspected that the necking may happen inside of the nozzle that is not discoverable with observation technique and the Young-Laplace equation (Fig. 7). As can be seen in this figure, the necking process is not observable in a non-Newtonian fluid, while it is clear in Newtonian one.

### 3.2. Force analysis

Utilizing the previously described force balancing, it is possible to approximate the vertical forces acting on the bubble during its evolution. They are plotted as a function of time in the nominal flow rate of 0.5 ml/min and 450 mm nozzle diameter as a sample in different continuous phases in Fig. 16. The results are qualitatively similar to the force evolution trends of Vafaei et al. [9] and Bari and Robinson [4] for their quiescent bubble growth test case. As can be seen, the trend of the changing procedure of the effective forces is similar in all liquids, independent of Newtonian/non-Newtonian behavior and concentration of the CMC solutions. It should be noted that the forces are just plotted during the expansion and detachment stages since there is no bubble and consequently no effective forces acting on the bubble during its waiting time.

It is clear that since the volume of the bubble increases continuously by approaching the detachment moment, the buoyancy force also increases and its value will be significant during the bubble elongation and departure stages.

The upwardly directed contact pressure force due to its dependency on the differential pressure is maximum near the beginning of the bubble growth when its shape is a kind of hemispherical and the pressure difference is maximum [4]. Following the trend of the gas pressure evolution, this force monotonically decreases as the time approaches the detachment moment.

The gas momentum force is another force considered as the upwardly directed forces which tends to detach the bubble from the nozzle rim. However, as can be seen in Fig. 16, its amount is negligible in comparison with other forces. That was predictable since the gas velocity is very low in this relatively low gas flow rates and as a result, the gas momentum does not affect the bubble evolution significantly in any stages of the formation.

The value of the hydrostatic force is also very small in all continuous phases, however, it has a larger contribution than the gas momentum force. This is in accordance with the previous observations in which for small flow rates the change in the hydrostatic head does not significantly affect the gas flow into the bubble [34].

The most important difference in the contribution of the effective forces is arising from the drag force, which is directly connected to the viscosity of the continuous phase. It can be seen that this force has the least contribution after the gas momentum and hydrostatic forces. However, its contribution generally increases with the incremental of the concentration of continuous phase. The difference is clear in Fig. 18, in which a noticeable increase in the contribution of drag force can be seen from water to 0.1% wt CMC solution. The increase is more obvious from 0.1% wt CMC to 0.25% wt CMC solutions, while it is almost constant as the concentration of the solution increases more (0.5% wt CMC solution). Some fluctuations can be seen in the amount of drag force, especially in high concentration solutions. These fluctuations can be justified by this fact that the bubble evolution has a bouncing behavior. This bouncing leads to the different projected areas and different shear rates (in non-Newtonian fluids) which consequently changes the drag force during the bubble evolution.

In order to verify the force balancing employed in this research, the net force is calculated also and is shown in all figures. The results show that in all continuous phases the net force is near zero, therefore, the assumptions made in the force balancing section are acceptable.

For a better understanding of the amount of the effective forces in different orifice diameters, Fig. 17 has been plotted, which shows a comparison of each mentioned forces contribution near detachment moment in distilled water at a nominal gas flow rate of 0.2 ml/min and different nozzle diameters. As can be seen, buoyancy and surface tension forces are more dominant that the other forces in all orifice
In more detailed observation it can be inferred that contribution of the surface tension force decreases as the orifice diameter increases. Contradictory behavior can be observed for buoyancy force, i.e., the influence of this force increases as the orifice diameter increases. This behavior can be justified using “square-cube” law: in general, forces ($f$) that are a function of area ($A$) of interaction decrease more slowly than properties that depend on the volume ($V$) [35]:

$$\frac{f(A)}{f(V)} \propto \frac{L^2}{L^3} \propto \frac{1}{L} \quad (21)$$

Fig. 16. The contribution of effective forces acting on the bubble near detachment moment in (a) water, (b) 0.1% wt CMC, (c) 0.25% wt CMC, and (d) 0.5% wt CMC as the continuous phase on 450 μm nozzle and 0.5 ml/min gas flow rate.

Fig. 17. The contribution of effective forces acting on the bubble near detachment moment versus nozzle diameter in distilled water and 0.2 ml/min gas flow rate.
where $L$ is the characteristic dimension of the microdevice. Based on this equation, the ratio of the surface forces to the volume forces varies from about $7 \times 10^4$ to $2 \times 10^4$. Therefore, in the smallest orifice diameter, surface tension has a larger contribution than the buoyancy force, while as the orifice diameter increases the buoyancy surpasses the surface tension as far as in 600 μm orifice diameter the buoyancy dominates the surface tension. Also, it can be seen that more than 90% portion of the forces belongs to the surface tension and buoyancy forces in all orifice diameters.

On the other hand, Fig. 18 shows a comparison of effective force contribution in different continuous phases. Obviously, buoyancy and surface tension have more contribution in distilled water, consequently, pressure and drag forces have less contribution in this fluid (meanwhile the influence of gas momentum force is negligible). Besides, Figs. 18 and 19 represent that trend of surface tension and pressure force are similar, i.e. with increasing viscosity and concentration of the continues phase, contribution of surface tension and pressure forces decrease. In contrast, the contribution of buoyancy and drag forces increase with increasing viscosity. Also, it can be inferred from Fig. 19 that the influence of hydrostatic force is not significant in all continues phases. However, its contribution relatively decreases as the viscosity of the continues phase increases. Hence, it can be concluded that in a high concentration of the continues phase, the drag force must be considered as an effective force acting on the bubble, while the hydrostatic force can be neglected. Whereas, in low viscosity fluids the drag force can be neglected and the hydrostatic force must be taken into account.

4. Conclusion

This research studied the formation of bubbles on different micro-meter-sized nozzles in Newtonian/non-Newtonian fluids under relatively low flow rate conditions ($0.1 \sim 1$ ml/min). Using high-speed camera and image processing technique and employing the Young-Laplace equation, detailed characteristics of the bubble formation during its evolution and especially near detachment moment were measured and analyzed. The major conclusions are as the following:

- Bubble volume obeys a U-shaped trend with increasing gas flow rate in both Newtonian/non-Newtonian fluids, however, is a weak function of gas flow rate.
- There is a critical flow rate in which bubbles formed on 600 μm nozzle has a lower volume than those formed on 450 μm nozzle. This critical flow is being delayed in non-Newtonian (shear-thinning) fluids, and also increases with increasing concentration of the continuous phase.
- The frequency of bubble formation changes linearly for a Newtonian
fluid, while a second-degree polynomial equation better fits data for non-Newtonian fluids in relatively low flow rates.

- Variation of the instantaneous contact angle with time shows a characteristic U-shaped trend, which affects bubble formation significantly through applying surface tension force.

- By applying the Young-Laplace equation, it was found that the necking process does not observable in non-Newtonian fluids since it happens somewhere inside the nozzle. Hence, the contact angle near detachment moment is less than 90° in these fluids.

- A force balance during bubble evolution reveals that buoyancy and surface tension forces are the most effective upward and downward forces, respectively.

- It was observed that drag force has noticeable contribution at a higher concentration of the CMC solution, while the hydrostatic force influence decrease with increasing of the concentration. Besides, it was concluded that the gas momentum is negligible in all fluids in such relatively low flow rates.

- It was shown that the micrometer-sized scale of the nozzles significantly influences the bubble formation process based on the "cube-squared law", i.e., the contribution of surface tension force increases as the nozzle diameter decrease.

References


[19] A. Terasaka, H. Tsuge, Bubble formation at a single orifice in Non-newtonian li-


[31] R. Darby, Transient and steady State rheological properties of very dilute drag re-


