Dynamics of a Single Air Bubble Rising in a Thin Gap Filled with Magnetic Fluids

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Abstract: The behavior of a single air bubble rising in magnetic fluids without magnetic fields is visualized using a mini-gap between two parallel vertical glass plates. Water-based Fe₃O₄ magnetic fluids with particle volume concentration of 6.33 % and density 1261.96 kgm⁻³ are filled in these gaps and a single air bubble is produced through the orifice at the bottom of the gap. The thicknesses of the gaps are 1 and 2 mm, respectively. Diameters of the orifices are 0.64 and 1.02 mm for 1 mm gap, and 0.64, 1.02 and 1.6 mm for 2 mm gap. In addition, four working fluids including pure water, 25% tetramethylammonium hydroxide (TMAH) aqueous solution and mass concentration 30% and 50% sucrose solution also have been tested. The results show that the shape of the bubble in magnetic fluids initially keeps oblate ellipse like that in the water, but after the middle of the journey, it becomes as an oblate elliptical cap like that in the surfactant aqueous solution. A reasonable explanation is obtained considering the diffusion of the surfactants to the interface of the bubble. Rise velocity of the bubble in magnetic fluids increases with the width of the gap, and the rise motion of the bubble become instable in the 2 mm gap, even breakup of a bubble generated by 0.64 mm orifice occurs.

Keywords: Magnetic fluids, bubble dynamics, surfactant.

1 Introduction

Magnetic fluids are stable colloidal dispersions containing single-domain magnetic particles with a size of about 10 nm in diameter [Rosensweig (1985)]. These particles are usually magnetite and are held in suspension by the use of surfactants which are compatible with both the carrier fluid and the particles. During the last few decades, researches of magnetic fluids have been extended to areas of physics, chemistry, engineering, and even medicine [Berkovsky, Medvedev and Krakov (1993); Berkovski and Bashtovoy (1996)]. Some novel applications have

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been proposed, such as micro/nano- electromechanical sensors, actuators, and micro/nanofluidic devices [Rinaldi, Franklin, Zahn and Cader (2004)], and targeted drug-delivery vectors in biomedical applications [Ganguly, Gaind, Sen and Puri (2005)].

Free interfacial flow of magnetic fluids under external magnetic fields becomes more complicated, since a jump in magnetic properties would found at fluid interfaces besides density and viscosity. Notable cases in point are normal field instability or Rosensweig instability in which an initially flat magnetic fluid surface exhibits peaked structure in a vertical magnetic field [Rosensweig (1985)], and labyrinthine patterns of a magnetic fluid drop formed in a Hele-Shaw cell [Dickstein, Erramilli, Goldstain, Jackson and Langer (1993)].

Due to the effect of the magnetic body force, the rise process of a single air bubble in magnetic fluids is quite different from that in common liquids. Generally, an applied magnetic field, even if stationary and uniform, can obviously modify the shape, trajectory and rise velocity. The bubble will be elongated along the direction of magnetic field, accelerated in the region of negative field gradient, and decelerated in the region of positive field gradient. Based on these characteristics, Kamiyama, Kamiya and Izu (1991) contrived an energy conversion device using bubbly flow produced by vaporization of the magnetic fluid due to heat addition. A large driving force will be generated in this system to push the magnetic fluid flow when a nonuniform magnetic field was introduced.

There are some papers have reported on the behavior of bubbles rising in magnetic fluids under different type of magnetic fields, experimentally and theoretically. Ishimoto, Okubo, Kamiyama and Higashitani (1995) observed the rising process of a single air bubble through a Hele-Shaw cell under a nonuniform magnetic field. They found that the shape of the bubble elongated along the field lines, and the rising velocity increases in the negative gradient area and decreases in the opposite case. The vapor bubbles generated in a vertical pipe also were investigated by using ultrasonic wave echo technique. Unfortunately, these images are too indistinct to identify. Subsequently, Ishimoto and Kamiyama (1996) numerically calculated the growth process and rise motion of a single bubble in Poiseuille flow taking into account the effect of magnetic body force made by a nonuniform magnetic field. The simulation verified their foregoing experimental results. Bashtovoi, Kovalev and Reks (2005a) observed an interesting phenomenon that a bubble cluster divided into two clusters in the presence of a uniform magnetic field, and the disintegration of the clusters enhanced when the field intensity increases. Then, they made another experimental investigation on air bubbles separation in magnetic fluids from the solid surfaces with the presence of uniform and nonuniform magnetic fields [Bashtovoi, Kovalev and Reks (2005b)]. The critical volume of bubbles

for separation would decrease more than 10 times, when the magnetic field gradient reached $1.5 \times 10^3 kAm^{-2}$.

Attention in the present work is focused on the effect of microstructure of magnetic fluids on the rising motion of a single bubble, and no field effect has been considered. It is well known that when the water contains surfactant additives, the velocity of the bubble rising in it is smaller than that in pure water at the same conditions, and the value of velocity will decrease to a minimum as the surfactant concentration increases [Clift, Grace and Weber (1978)]. The physical mechanism was first explained by Levich (1962), what the reason is the surface tension gradient caused by surfactants. Owing to the effect of diffusive and convective mass transport, the surfactant concentration at the rear of the bubble is greater than that at the front, and the resulting Marangoni tension needs a viscous shear stress on the bubble as compensation, which will decrease the rise velocity [Fdhila and Duineveld (1996)]. More detailed discussions can be found in these literatures [Bush (1997); Liao and McLaughlin (2000); Zhang and Finch (2001); Hestroni, Mosyak, Pogrebnyak, Sher and Segal (2006)].

The surfactant additives in magnetic fluids are comprised of two parts: the molecules absorbed on the nanoparticles and those which dissociated in the carrier liquid. The previous researches treated the magnetic fluid as a homogeneous liquid with a certain magnetization, and no influence of internal configuration has been taken into account for the study of bubble behavior in magnetic fluids. As we know, the influence of suspended magnetic nanoparticles on the shape and trajectory of the bubble keeps unclear. The macroscopic effect of the nanoparticles is mainly increasing the "effective viscosity" of the colloid [Odenbach (2002)].

The objective of this work is to understand the behavior of a single air bubble rising in magnetic fluids, especially to study the influence of the surfactant additives and the magnetic nanoparticles on the shape, trajectory and rise velocity of the bubble. A visualization experiment was conducted when a bubble formed in the narrow gap between two parallel vertical glass plates (like a Hele-Shaw cell). The results were compared with those in pure water, surfactant aqueous solution, and sucrose solution.

2 Experimental

Since the magnetic fluid is black and opaque, it is impossible to observe the bubbles rising through extended magnetic fluid by naked eye. Therefore, the visualization was realized using a device which is similar to a vertical Hele-Shaw cell consisted of two parallel glass plates. The bubble formed at the bottom rises through the gap, and the profile of the bubble can be captured under a background light. Extreme

cautions were taken to keep the plates as clean as possible.

A water-based Fe₃O₄ magnetic fluid with particle volume concentration of 6.33% and density 1261.96 kgm⁻³ was used as working fluid, which was prepared by coprecipitation method using tetramethylammonium hydroxide (TMAH) aqueous solution as surfactants. At the same time, pure water, mass concentration 25% TMAH aqueous solution, and mass concentration 30% and 50% sucrose aqueous solution also have been filled in the gaps. The shapes, trajectories and rise velocities of the bubbles rising in these liquids are compared with each other.



Figure 1: Schematic diagram of the experimental apparatus

The schematic diagram of the experimental apparatus is shown in Fig. 1. The narrow gaps are made of two parallel vertical glass plates with dimensions of 121×120 mm, and the thicknesses of the gaps are 1 and 2 mm, respectively. A single air bubble is produced through the orifice at the bottom of the gap by promoting the syringe which is connected to the orifice. Diameters of the orifices are 0.64 and 1.02 mm

for 1 mm gap, and 0.64, 1.02 and 1.6 mm for 2 mm gap. The height of the fluids in the gaps is nearly 120 mm. Pressure in the syringe was measured by a pressure transducer with uncertainty of 0.075% (Rosemount 3051 GP series) so that the state of the air before forming bubble could be confirmed. Three T-type thermocouples were disposed to measure the temperatures of working fluids, environment and the air in the connecting pipe. These data were collected by a data acquisition system (IMP 3595 series), and the analog were converted to digital data for storing in computer. Experiments were carried out under conditions of atmospheric pressure (0.097MPa) and room temperature of 25° .

A CCD camera was employed to visualize bubble behavior in the gaps. For a typical experimental run, the rise motion of air bubble released at the bottom of the cell were recorded at a speed of 25 pictures per second, and the time between two consecutive pictures was 40 ms. The bubbles were simplified as two-dimensional, and no 3D effects were considered. The image contained 350×280 pixels and typically covered an area of 120 mm \times 91 mm. During the image analysis, the centroid and outline of the bubbles were captured by the software of SigmaScan[®], so that the rise velocity could be calculated by dividing the time interval at a given experimental condition.

3 Result and discussion

3.1 Shapes and trajectories

Figure 2 and Fig. 3 show the superimposed frames of a single bubble from the detachment from the orifice to the annihilation at the free surface of the liquids. The bubbles are generated by 0.64 mm and 1.02 mm orifices in a 1 mm gap. In these figures, the MF denotes magnetic fluids, TMAH denotes mass concentration 25% tetramethyl- ammonium hydroxide aqueous solution, and Sucrose 30% and Sucrose 50% denote mass concentration 30% and 50% sucrose aqueous solution, respectively. The time in the bracket indicates the time interval of the adjacent bubbles. When air was continuously injected into the gap at the lower central orifice by promoting the syringe a bubble began to form and grow. As it did so, it detached from the bottom, and rose as a self-contained entity under the action of the buoyancy force acting upon it.

The diameters *d* of bubbles rising in the 1 mm gap are in range of 5-10 mm, which are much larger than the gap thickness $b (d/b \gg 1)$, so a two-dimensional description is receivable. All bubbles in five working fluids followed a straight line except pure water, in which the breakup of the bubble generated by 0.64 mm occurred. The trajectory of the bubble in water produced by 1.02 mm slightly deviated from the axis line when it approached the free surface, the reason may be the lower viscosity



Figure 2: Trajectories of the bubbles generated by 0.64 mm orifice rising in 1 mm gap



Figure 3: Trajectories of the bubbles generated by 1.02 mm orifice rising in 1 mm gap

of water.

The shape of the bubble rising in magnetic fluids initially kept oblate elliptical like that in the water and 30% sucrose solution, but after middle of the journey, it became as an oblate elliptical cap like that in the surfactant aqueous solution. The curvature at the nose of bubble was obviously larger than that at the rear, while the bubble in water always kept same shape in the whole process. The case in the surfactant solution was completely different from that in the water, where the bubble exhibits approximately spherical cap with its symmetry axis parallel to the bubblecentre velocity, and the bubble shape remains constant and slightly elongated along the vertical axis after the initial acceleration stage. This phenomenon could be explained as the existence of surfactant additives. As the bubble rose in the liquid, surfactants tend to adhere to the bubble interface, which will result in a lower, non-uniform surface tension along the interface and introduce the Marangoni force. Further, there may be exchange (adsorption/desorption) of surfactants between the interface and the bulk. As a result, the concentration of surfactant additives on the bubble interface is nonuniform. Since the velocity of bubble is larger than the surrounding liquid, surfactants will be accumulated at the trailing end of the bubble, and a surfactant concentration difference between the front and back of bubble will be produced. Then, the deformation of lower half of the bubble surface was restricted, and the bending of the upper half became relatively easy. The content of the surfactant additives is small, and most of them are coated on the magnetic nanoparticles, only a few loitered in the carried fluid. There is a relaxation when the accumulation of dissociative surfactants on the rear of bubble achieved to the extent which can make influence on the appearance of the bubble, so the behavior of bubble in magnetic fluids only takes the shape of that in TMAH aqueous solution at the remaining half of journey.

From these pictures, we can also conclude that the behavior of bubble in magnetic fluids is similar to that in the mass concentration 30% sucrose aqueous solution at the initial stage. The viscosity of magnetic fluids is larger than pure water due to the influence of nanoparticles. The viscosity will increase to 1.45 mPa·s for magnetic fluids with particle volume concentration of 6.33%, which can be calculated from the Rosensweig's formula [Rosensweig (1985)] in no magnetic field case:

$$\eta = \eta_0 \left(1 - \frac{5}{2}\varphi + \left(\frac{5}{2}\varphi_c - 1\right) \left(\frac{\varphi}{\varphi_c}\right)^2 \right)^{-1}$$
(1)

Where η is the dynamic viscosity of magnetic fluids, η_0 is the viscosity of carrier liquid, ϕ the volume concentration of particles including their surfactant, ϕ_c the critical volume concentration, usually be 0.74. And the viscosity of 30% sucrose solution is 2.78 mPa·s [Mathlouthi and Gotelle (1995)], twice as the magnetic fluids, and with the same value of the surfactant solution (about 2.8 mPa·s). The deformation of the bubbles in the 50% sucrose solution is not significant. They remain spherical and their trajectory is rectilinear. They have effective diameters less than 6 mm, nearly half of the magnetic fluids, which may be caused by their higher density of 1227.37 kg/m³ [Bubnik, Kadlek, Urban and Bruhns (1995)] and viscosity of 12.6 mPa·s[Mathlouthi and Gotelle (1995)]. The larger density difference between the working fluids and air would result in the reduction of the bubble departure diameter. Further, the smaller the size of the bubble was, the deformation

of the bubble was less obvious, and the bubble kept sphere easier. In addition, the viscous drag force also resisted the expansion of the bubble surface.



 Water
 MF
 TMAH Sucrose 30% Sucrose 50%

 (40 ms)
 (40 ms)
 (80 ms)
 (40 ms)

Figure 4: Trajectories of the bubbles generated by 0.64 mm orifice rising in 2 mm gap



Figure 5: Trajectories of the bubbles generated by 1.02 mm orifice rising in 2 mm gap

Figure 4 to Fig. 6 show the rise motion of bubbles through the 2 mm gap, where the bubbles were produced by 0.64, 1.02 and 1.6 mm orifices. From the pictures, the shapes of the bubbles varied obviously from each other as the diameter of the orifice increases, even breakups of bubbles in magnetic fluids and 30% sucrose





solution occurred when the diameters of the orifices are 0.64 and 1.02 mm. The bubble diameters increase substantially compared to the 1 mm gaps at the same condition, especially which of the 50% sucrose solution increases almost double. And the diameter of the bubble generated by the 1.02 mm orifice in magnetic fluids even reached 12 mm.

A plane zigzag trajectory can be found in water, magnetic fluids and even 30% sucrose solution, while the trajectories of bubbles in surfactant and 50% sucrose solution still keep rectilinear. The onset of the path instability was delayed when bubbles generated through small orifices and these bubbles have bigger diameters. But the oscillation deviated from the centerline started as song as the bubble detached from the orifice for the case of 1.6 mm orifice, and the diameter was smaller than the previous examples. Kelley and Wu's experimental results (1997) indicated that the path instability in a Hele-Shaw cell was caused by the vortex shedding in the wake of the bubble.

Similar to the behavior of the bubble in 1 mm gap, the shape of the bubble in magnetic fluids transformed form an oblate ellipse to an oblate elliptical cap after the middle of the journey. Small orifice may produce big bubble, even the bubble produced by the 0.64 mm orifice burst after a distance from the bottom. The trajectory of the bubble produced by the 1.6 mm orifice initially kept rising in a straight line, but the path became instable at the late stage of the rise motion. The bubbles in magnetic fluids and 30% sucrose solution have almost the same trajectories.

Compared to the 1 mm gap, the wall effect is not so significant for the 2 mm gap. When the departure diameter reduced to the order of the gap thickness, such as the situation in the Fig. 6, the motion of bubbles became instable considerably like the situations in the bulk liquid.

3.2 Rise velocities

The rise motion of the bubbles is determined by buoyancy, surface tension, viscous drag force and so on. Therefore, the rise velocities of the bubbles are strongly influenced by the properties of the fluids. Fig. 7 and Fig. 8 show the typical velocity of a single bubble as a function of *y* coordinate, where the bubbles in Fig.7 were generated by 1.02 mm orifice rising in 1 mm gap and in Fig.8 were generated by 1.6 mm orifice rising in 2 mm gap, respectively. The transient velocities were given by the distance between the centroid of two adjacent bubbles dividing by the time interval, which is a constant of 40 ms. The initial data point was set as the centroid of the bubble which completely detached the orifice. The whole process of the rise motion could be divided into three stages: acceleration process with positive velocity gradient, free rise with constant velocity, and annihilation stage where the bubble dramatically decelerated and finally burst at the surface of the liquid. In general speaking, the terminal velocity is the velocity in free rise period.



Figure 7: Velocities of the bubbles generated by 1.02 mm orifice rising in 2 mm gap

According to the values of the viscosities of the working fluids, the largest one was 50% sucrose solution followed in descending order by surfactant solution, 30% sucrose solution, magnetic fluids, and water. The terminal velocities of the bubbles



Figure 8: Velocities of the bubbles generated by 1.6 mm orifice rising in 1 mm gap



Figure 9: Velocities of the bubbles rising in magnetic fluids

rising in water and magnetic fluids have larger values, and the 50% sucrose solution has the smallest value, which are inversely proportional to their viscosities. The viscous drag force plays an important role on the bubble rising in a narrow gap. The abnormal phenomenon happened on the surfactant solution, the rise velocity

of it was smaller than that in 30% sucrose solution although they have nearly same values of viscosities, even smaller slightly than the situation in 50% sucrose at the later period. It seemed that the surfactant additives decreased the velocity as described in the paper of Fdhila and Duineveld (1996). The surfactants induced additional shear stress which resisted the rise motion of the bubble.

Moreover, when the gaps were filled with other fluids except 50% sucrose solution, the velocities of the bubbles in 2 mm gap were larger than those in 1 mm gap. It can be concluded that the wall effect has an obvious influence on the lower viscous fluids. Compared with the bubble in water, the fluctuation of the velocity of the bubble in 50% sucrose solution was negligible, and the bubble in it kept a uniform speed. The velocity of the bubble rising in magnetic fluids was smaller than that in water in 1 mm gap, but they were almost the same when the bubbles rose in 2 mm gap.

Figure 9 shows the velocities of the bubbles rising in magnetic fluids. It was observed that the velocities of the bubbles in 2 mm gap were much larger than those in 1 mm gap, even the velocity of the bubble generate by 1.02 mm orifice was double of that in 1 mm gap. The velocities of the bubbles generated by 0.64 mm and 1.02 mm orifices in 1 mm gap were almost the same. The bubble always kept a uniform speed till it reached the free surface where the speed having a significant decrease. On the other hand, the velocities of the bubbles rising in 2 mm gap were instable, and the bubble generated by 0.64 mm orifice broken up occurred at the middle of the journey. Though the rise motion of the bubble generated by 1.02 mm orifice.

4 Conclusions

The rise motions of a single bubble rising in magnetic fluids and other four working fluids in narrow gaps were studied experimentally. Owing to the existence of the magnetic nanoparticles, the viscosity of the magnetic fluid was larger than that of the carried liquid, and the larger viscous drag force was induced. Additionally, the rise motion of the bubble also was affected by the dissociative surfactants. As a result, the behavior of the bubble rising in magnetic fluid becomes more complicated. Under the influence of surfactants, the shape of the bubble rising in magnetic fluids will transform from an oblate ellipse to an oblate elliptical cap after the initial acceleration stage. Furthermore, the rise velocity was smaller than that in water in 1 mm gap due to the larger viscous drag force, and the bubbles rose along the vertical axis with a uniform speed. But in the 2 mm gap case, the shapes and paths became irregular, even breakup and zigzag trajectory appeared. The path of bubble in magnetic fluids still kept rectilinear at the initial stage due to its bigger viscosity, meanwhile the bubble immediately swung in water after the detachment from the

orifice.

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