THEORETICAL AND EXPERIMENTAL STUDIES OF THE DEFORMATION OF BUBBLES MOVING IN QUIESCENT NEWTONIAN AND NON-NEWTONIAN LIQUIDS

TAKAO TSUKADA, HARUYUKI MIKAMI AND MITSUNORI HOZAWA

*Chemical Research Institute of Non-Aqueous Solutions,
Tohoku University, Sendai 980*

NOBUYUKI IMAISHI

Institute of Advanced Material Study, Kyushu University, Kasuga 816

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Theoretical and experimental studies of the shapes and terminal velocities of bubbles rising in quiescent Newtonian and non-Newtonian liquids were carried out. For non-Newtonian liquids, the power law model was adopted as the constitutive model.

Numerical analysis by use of the finite element method can predict well the observed shapes and velocities of such bubbles.

It is found experimentally and theoretically that as bubble diameter increases, the bubble deforms from a sphere to one of three types of ellipsoids, depending on the viscosity of the continuous phase: (1) more deformed at the front than at the rear, (2) symmetrical front to rear, and (3) more deformed at the rear than at the front, in both Newtonian and non-Newtonian liquids.

Shape regime maps are proposed on the basis of the numerical simulations.

Introduction

As a correct knowledge of the behavior of bubbles rising in a quiescent liquid is of practical importance in the design and operation of gas-liquid contact mass transfer equipment, a number of experimental studies have been carried out. For bubbles in Newtonian liquids, Grace²⁾ has brought together the data from different investigators and produced a dimensionless diagram of the terminal velocity and the shape as a function of bubble size for a wide range of physical properties. Bhaga *et al.*¹⁾ extended Grace's work and presented diagram for bubbles with many kinds of shape regimes. With the advances of polymer technology and biotechnology, rheological studies of bubbles have been carried out experimentally. For example, experimental work on the motion of dissolving and non-dissolving bubbles in viscoelastic liquids was reported by Zana *et al.*⁹⁾

Theoretical approaches^{3-5,7)} to the above problems have also been made. However, most of the works have investigated the cases of bubbles slightly deformed from spherical shape.

Recently, Ryskin *et al.*⁶⁾ obtained numerical results for the motion of a deformed bubble rising through a quiescent Newtonian liquid, using the finite difference method and the boundary-fitted, orthogonal coordinates technique. The authors⁸⁾ also sim-

ulated numerically the deformations of moving bubble and drop in a Newtonian liquid by use of the finite element method. However, there has been no report regarding the deformation of a bubble in a non-Newtonian liquid.

In this work, the mathematical model in our previous work⁸⁾ was extended to deal with the motion of a bubble which rises through a non-Newtonian liquid and is more deformable than that in the previous work involving both Newtonian and non-Newtonian liquids. The results obtained were compared with experimental ones to verify the mathematical model. Diagrams of the bubble shapes for both liquids are also proposed, based on the results of the numerical simulations.

1. Theory

Figure 1 shows the cylindrical coordinates. A single bubble rising in a quiescent Newtonian or non-Newtonian liquid is considered.

The following assumptions are used in the calculations. (1) The system is in steady state and is axisymmetric. (2) The motion inside the bubble does not affect the flow of the continuous phase. Under the above assumptions, the governing equations, i.e. momentum and continuity equations, for the continuous phase are given as follows.

$$-\nabla \cdot \rho \mathbf{v} \mathbf{v} - \nabla p - \nabla \cdot \boldsymbol{\tau} + \rho \mathbf{g} = 0 \quad (1)$$

$$\nabla \cdot \mathbf{v} = 0 \quad (2)$$

* Received 1989. Correspondence concerning this article should be addressed to T. Tsukada. H. Mikami is now with Sumitomo 3M Ltd., Sagami-hara 229.

Fig. 1. Cylindrical and spherical coordinate systems

The boundary conditions are expressed by the following equations.

$$\text{At inflow plane: } u=0, \quad v=U \quad (3-1, 2)$$

$$\text{At outflow plane: } u=0, \quad \partial v / \partial z = 0 \quad (3-3, 4)$$

At outside wall boundary:

$$u=0, \quad v=U \quad (3-5, 6)$$

At bubble surface:

$$\mathbf{n} \cdot \mathbf{v} = 0 \quad (3-7)$$

$$\boldsymbol{\tau} : \mathbf{n}\mathbf{n} = 0 \quad (3-8)$$

where U indicates the terminal velocity of a single bubble.

For a non-Newtonian liquid, the power law model is used as the constitutive equation, being defined by the following equations.

$$\boldsymbol{\tau} = -2\eta(I_2)\mathbf{d} \quad (4)$$

where

$$\mathbf{d} = (\nabla \mathbf{v} + \nabla \mathbf{v}^T) / 2 \quad (5-1)$$

$$\eta(I_2) = \kappa I_2^{n-1} \quad (5-2)$$

$$I_2 = (2\mathbf{d} : \mathbf{d})^{1/2} \quad (5-3)$$

in which $\mathbf{d}$ is the rate of deformation tensor and I_2 is the second invariant of $\mathbf{d}$. In Eq. (5-2), if $n=1$ and $\kappa=\mu$ the fluid is Newtonian, and the deviation of n from unity indicates the degree of non-Newtonian behavior. In this work, a fluid of $n<1$, i.e. a pseudoplastic liquid, was used.

The shape of the bubble surface is obtained by solving the equation for the normal stress balance on the bubble surface, Eq. (6), under the following constraint conditions, where the spherical coordinates (f, θ) are used as shown in Fig. 1.

$$p + \boldsymbol{\tau} : \mathbf{n}\mathbf{n} - 2H\sigma = p_G \quad (6)$$

The bubble volume is given by Eq. (7).

Fig. 2. Discretization of calculation domain for the finite element method. (Figure on the right is enlargement of elements around a bubble, that is, of the smallest circle in the figure on the left.)

$$(2/3)\pi \int_0^\pi f^3 \sin \theta d\theta = \pi d_e^3 / 6 \quad (7)$$

The origin of the coordinates is fixed at the mass center of the bubble, and then

$$\int_0^\pi f^4 \sin \theta \cos \theta d\theta = 0 \quad (8)$$

To solve this problem, the Galerkin finite element method is used. The calculation domain is discretized by isoparametric elements consisting of a nine-node quadrilateral as shown in Fig. 2, where the total number of elements and nodal points are 442 and 1855 respectively.

In each element, velocity vector $\mathbf{v}$ and pressure p are approximated with the biquadratic and bilinear trial functions, respectively.

$$\mathbf{v}(r, z) = \sum_i \phi_i(r, z) \mathbf{v}_i \quad (9)$$

$$p(r, z) = \sum_k \psi_k(r, z) p_k \quad (10)$$

The bubble shape is expressed by Eq. (11).

$$f(\theta) = \sum_i \chi_i(\theta) f_i \quad (11)$$

By substituting Eqs. (9)–(11) into Eqs. (1), (2) and (6), the Galerkin procedure provides a set of algebraic equations. The equations are solved by use of the Newton-Raphson method.

2. Experimental

Figure 3 shows a schematic diagram of the apparatus.

A single bubble is formed, using a turning cup of acrylic resin at the bottom of a Pyrex glass column (135 cm high and 13 cm in diameter). After release

Fig. 3. Schematic diagram of apparatus: 1. test column; 2. laser beam source; 4, 5. photo transistor; 6. flash lamp; 7. camera; 8. pulse generator; 9. electronic timer; 10. motor drive unit; 11. turning cup; 12. needle; 13. mercury

from the turning cup, the bubble rises and cuts the two laser beams sequentially. Pulses from the photo-transistors start and stop the electronic timer. The terminal velocity of the bubble is determined by the distance and the elapsed time between the two laser beams. When the bubble passes the lower beam, a pulse activates a camera and a flash lamp, to photograph the bubble. The test section (between the two beams) is placed 50 cm above the turning cup to ensure steady-state motion of the bubble. The volume and the volume equivalent diameter of the bubble can be determined from the photograph.

Aqueous glycerol solutions and aqueous CMC (carboxy methyl cellulose sodium salt) solutions of different concentrations were used as the Newtonian and non-Newtonian liquids, respectively. For the non-Newtonian liquids, the rheological parameters, i.e. n and κ in Eq. (4), were measured by using a cone-and-plate viscometer. Air or nitrogen was used as the gas phase.

The temperature was kept at $23 \pm 1^\circ\text{C}$ during the experiments.

3. Results and Discussion

3.1 A single bubble rising in Newtonian fluid

Figure 4 shows the relationship between the terminal velocity U and the volume equivalent diameter d_e for bubbles rising in aqueous glycerin solutions with three different concentrations. The viscosity of each solution is shown in Fig. 4. In this figure, the lines and the symbols show the calculated and the experimental values, respectively. For the experimental datum expressed by the open square, a zigzag motion of the bubble was observed. The calculated results in the present analysis are all in good agreement with the experimental ones.

The d_e - U relationship in Fig. 4 can be transformed into the relationship between Reynolds number Re

Fig. 4. Terminal velocity U vs. volume equivalent diameter d_e for bubbles in aqueous glycerin solutions

Fig. 5. Drag coefficient C_D vs. Reynolds number Re for bubbles in aqueous glycerin solutions

and drag coefficient C_D shown in Fig. 5. The solid line of $C_D = 16/Re$ in Fig. 5 is the Hadamard-Rybczynski analytical solution for creeping flow. Since the calculated results agree well with the experimental ones and coincide with the analytical solution for $Re < 1$, the present calculation method is reasonable.

Figure 6 shows the aspect ratio of a bubble, a/b , as a function of volume equivalent diameter d_e , where a and b indicate the minor and the major axes of a bubble respectively if the bubble shape is ellipsoidal. From the calculated and experimental results for the three solutions, it is found that the bubble deforms from a sphere ($a/b=1$) to an oblate ellipsoid as d_e increases. For large d_e bubbles in the liquid of the highest viscosity, the observed ratio a/b approaches a constant value corresponding to a spherical cap.

Figure 7 shows the velocity vectors and the shapes of bubbles at different Reynolds numbers, calculated

Fig. 6. Aspect ratio of bubbles a/b vs. volume equivalent diameter d_e for bubbles in aqueous glycerin solutions

Fig. 7. Calculated velocity vectors and shapes of rising bubbles in aqueous glycerin solutions:

- (1) $\mu=0.004$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $M=9.99 \times 10^{-9}$
- (2) $\mu=0.029$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $M=2.04 \times 10^{-5}$
- (3) $\mu=0.110$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $M=4.08 \times 10^{-3}$

for three concentrations of glycerine, that is, for three viscosities. For the three cases, the deviation from spherical shape increases as Re increases, although the deformation for large Re is classified into three types as follows: (1) in low-viscosity liquid the bubble deforms more at the front than at the rear; (2) in intermediate-viscosity liquid the bubble shape is symmetrical front to rear; and (3) in high-viscosity liquid the bubble deforms more at the rear than at

Fig. 8. Experimental bubble shapes rising in aqueous glycerin solutions

- (1) $Re=189.3$, $\mu=0.004$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $M=9.99 \times 10^{-9}$
- (2) $Re=73.8$, $\mu=0.029$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $M=2.04 \times 10^{-5}$
- (3) $Re=24.6$, $\mu=0.110$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $M=4.08 \times 10^{-3}$

Fig. 9. Distributions of static (-----) and dynamic (.....) pressure, normal viscous stress (.....) and their sum (—) on the bubble surface in aqueous glycerin solutions

- (1) $\mu=0.004$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $Re=195.8$, $M=9.99 \times 10^{-9}$
- (2) $\mu=0.029$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $Re=79.50$, $M=2.04 \times 10^{-5}$
- (3) $\mu=0.110$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], $Re=23.49$, $M=4.08 \times 10^{-3}$

the front. These three types of deformation were also observed experimentally as shown in Fig. 8.

Figure 9 shows the distributions of static and

dynamic pressures, normal viscous stress and the their sum on the bubble surface for three viscosities in a dimensionless form, where $1/2\rho U^2$ is used as the characteristic pressure. The results of the sum of the pressure reveal the following: (1) For the bubble in low-viscosity liquid in Fig. 9(1), the front stagnation pressure becomes dominant. (2) For the bubble in the intermediate-viscosity liquid in Fig. 9(2), the front and rear stagnation pressures are almost the same. (3) For the bubble in high-viscosity liquid in Fig. 9(3), the rear stagnation pressure is dominant. As the bubble shape is determined by the normal stress balance on the bubble surface as expressed by Eq. (6), the bubble deforms to one of the three types of ellipsoids shown in Fig. 7 according to their pressure distributions on the bubble surface, which depend on the viscosity. Ryskin *et al.*⁶⁾ obtained the same results in their theoretical works. However, the reason why the pressure becomes dominant at the front of the bubble in low-viscosity liquid or at the rear in high-viscosity liquid is not evident.

So far, the relationships between U and d_e , and between a/b and d_e , have been discussed separately. Grace²⁾ suggested that the shape of a bubble can be uniquely mapped on a Re - Bo plane with the Morton number as a parameter. The dotted line in Fig. 10 indicates Grace's mapping, classifying the bubble shapes into sphericals, ellipsoidal and spherical caps. According to the present analysis, the bubble shape should be classified into three types as indicated by the hatched lines in Fig. 10: ① spherical bubble, defined as a bubble of $a/b > 0.9$ after Grace's definition²⁾; ② an ellipsoidal bubble which deforms more at the front; and ③ an ellipsoidal bubble which deforms more at the rear. The boader line between ② and ③ is drawn so that the distance of the major axis from the front stagnation point to its distance from the rear stagnation point is the same. In Fig. 10, the symbols show the experimental data for the aqueous glycerin solutions, where the triangular solid symbols indicate spherical bubbles and the rest indicate nonspherical ones.

3.2 A single bubble rising in non-Newtonian liquid

Figures 11 and 12 respectively show the relationships between U and d_e , and between Re and C_D , for a single bubble rising in aqueous CMC solution. The rheological parameters in the power law model, n and κ , are shown in Figs. 11 and 12. In both figures, the lines and the symbols respectively show calculated and experimental results. The analytical solutions by Kawase *et al.*³⁾ for creeping flow which obey the power law model are also shown in Fig. 12. The calculated results are in good agreement with the experimental ones, except for those in the case of small Re (or d_e). Since at small Re in Fig. 12 the experimental data approach the behavior of rigid spheres in a power-law

Fig. 10. Shape regime map for bubbles rising in Newtonian liquids: ① spherical bubbles, ② bubbles more deformable at the front, ③ bubbles more deformable at the rear.

Fig. 11. Terminal velocity U vs. volume equivalent diameter d_e for bubbles in aqueous CMC solutions

Fig. 12. Drag coefficient C_D vs. Reynolds number Re for bubbles in aqueous CMC solutions

Fig. 13. Aspect ratio of bubbles a/b vs. volume equivalent diameter d_e for bubbles in aqueous CMC solutions

fluid, the cause of the deviation of the experimental data from the calculated data may be attributed to the effect of contamination.

Figure 13 shows the relationship between a/b and d_e for a non-Newtonian liquid. The calculated results for the liquids with lowest and intermediate concentrations of CMC agree approximately with the experimental ones, although the experimental values of a/b show positive deviations from the calculated ones at small d_e . The bubble in the highest-concentration liquid, however, shows essentially different behavior, since it deforms from spherical to prolate teardrop shape ($a/b > 1.0$), and then to an oblate ellipsoid, as d_e increases. This behavior of bubbles is often observed in viscoelastic liquids, and the elongation in the direction of motion into teardrop shape is known to be caused by the effect of elasticity of liquids. Our experimental results suggest that the effect of elasticity becomes large in aqueous CMC solution if the concentration is high. However, this effect cannot be taken into account in the present analysis based on the power law model.

Figure 14 shows the calculated results for the velocity vectors and shapes of the bubble in CMC solutions of three different concentrations. The bubble shapes are classified into three types at large Re , almost the same as those in the Newtonian liquids. Typical bubble shapes in low- and medium-viscosity solutions are shown in Fig. 15, where the case of the highest concentration is eliminated because of the large deviation between calculations and experiments shown in Fig. 13. From Figs. 7 and 14 it is found that the bubble deforms from a sphere to one of three types of ellipsoids in both Newtonian and non-Newtonian liquids.

For non-Newtonian liquids, the shape regime map of bubbles is also provided in Fig. 16 by numerical simulations in the same manner as in Fig. 10, where n in the power law model is assumed to be 0.7. Only the boundary lines between the spherical bubbles and

Fig. 14. Calculated velocity vectors and deformations of rising bubbles in aqueous CMC solutions

- (1) $\kappa = 0.018$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{n-2}$], $n = 0.98$, $M = 2.11 \times 10^{-6}$
 (2) $\kappa = 0.083$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{n-2}$], $n = 0.90$, $M = 2.67 \times 10^{-4}$
 (3) $\kappa = 0.290$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{n-2}$], $n = 0.87$, $M = 2.46 \times 10^{-2}$

Fig. 15. Experimental bubble shapes rising in aqueous CMC solutions

- (1) $Re = 91.3$, $\kappa = 0.018$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{n-2}$],
 $n = 0.98$, $M = 2.11 \times 10^{-6}$
 (2) $Re = 41.9$, $\kappa = 0.083$ [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{n-2}$],
 $n = 0.90$, $M = 2.67 \times 10^{-4}$

the ellipsoidal bubbles are shown in Fig. 16. The boundary lines for both liquids are approximately coincident.

Conclusions

The deformations of single bubbles rising in both Newtonian and non-Newtonian liquids were investigated theoretically and experimentally, and the following conclusions were obtained.

- (1) The theoretical method presented here can predict the shapes and the terminal velocities of

Fig. 16. Shape regime map for bubbles rising in non-Newtonian liquids ($n=0.7$)

- ① spherical bubbles
② ellipsoidal bubbles

bubbles in Newtonian liquids and non-Newtonian liquids as described by the power law model.

(2) As bubble size increases, the bubbles deform from spheres to three types of ellipsoids according to the viscosity of the continuous phase, for both Newtonian and non-Newtonian liquids.

(3) Non-dimensional shape regime maps are presented for Newtonian and non-Newtonian liquids.

Nomenclature

a	= minor axis of ellipsoidal bubble	[m]
Bo	= Bond number ($= \rho g d_e^2 / \sigma$)	[—]
b	= major axis of ellipsoidal bubble	[—m]
C_D	= drag coefficient ($= 4gd_e / 3U^2$)	[—]
d	= rate of deformation	[s ⁻¹]
d_e	= volume equivalent diameter	[m]
f	= radial distance in spherical coordinates	[m]
g	= gravity acceleration	[m·s ⁻²]

H	= curvature	[m ⁻¹]
I_2	= second invariant of d	[m ⁻¹]
M	= Morton number ($= g^{3n-2} \kappa^4 / (\rho^{2-n} \sigma^{n+2})$)	[—]
n	= parameter in power law model	[—]
$\mathbf{n}$	= outward normal vector on boundary surface	[—]
p	= pressure	[Pa]
P_G	= pressure in gas bubble	[Pa]
Re	= Reynolds number ($= d_e^n U^{2-n} / \kappa$)	[—]
r	= radial distance in cylindrical coordinates	[m]
U	= terminal velocity of bubble	[m·s ⁻¹]
u	= radial component of velocity vector	[m·s ⁻¹]
$\mathbf{v}$	= velocity vector	[m·s ⁻¹]
v	= axial component of velocity vector	[m·s ⁻¹]
$\mathbf{t}$	= tangential vector on boundary surface	[—]
z	= axial distance in cylindrical coordinates	[m]
θ	= polar angle in spherical coordinates	[deg]
κ	= parameter in power law model	[kg·m ⁻¹ ·s ⁿ⁻²]
μ	= viscosity	[kg·m ⁻¹ ·s ⁻¹]
ρ	= density	[kg·m ⁻³]
σ	= surface tension	[N·m ⁻¹]
τ	= stress tensor	[Pa]
ϕ_i	= biquadratic trial function	[—]
ψ_k	= bilinear trial function	[—]
χ_l	= Hermit cubic trial function	[—]

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