Note

Reply to the Paper Named “The Force Acting on a Sphere Moving towards a Solidification Front due to an Interfacial Energy Gradient at the Sphere/Liquid Interface”

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1. Introduction

Our paper named “Behavior of fine bubbles in front of the solidifying interface”¹⁾ was published in ISIJ International (Vol. 39, 1999, p. 553). Recently, we received the comment on this paper from Kaptay and Kelemen.²⁾ Here are our replies to their comment.

2. Interpretation of Force Acting on a Sphere due to an Interfacial Tension Gradient

Kaptay *et al.*²⁾ attempted to derive the driving force from the aspect of energy. They gave the surface area of sphere particle, A , and the total interfacial energy, G , and the driving force, F_I , as:

$$G = \int_{A_b} \sigma dA \dots\dots\dots(1)$$

$$F_I = -\frac{dG}{dx} = -4\pi R^2 K \dots\dots\dots(2)$$

where $K (=d\sigma/dx)$ is the interfacial energy gradient or interfacial tension gradient, R is the radius of sphere particle.

On the other hand, Mukai and Lin³⁾ derived the driving force that acts on sphere particle located in liquid with an interfacial tension gradient and obtained Eq. (3).

$$F_I = -(8/3)\pi R^2 K \dots\dots\dots(3)$$

We can also derive F_I based on Laplace equation. Let's consider the following system (Fig. 1) in which only interfacial tension gradient force acts on the particle, for example, under the microgravity field. Laplace equation gives,

$$P_p = P_o + \frac{2\sigma}{R}$$

$$K = d\sigma/dz, \quad \sigma = \sigma_o + Kz,$$

$$dF_I = \left[P_o + \frac{2(\sigma_o + Kz)}{R} \right] df$$

$df = 2\pi R dz$ (surface area of ring AA'B'B in Fig. 1)

$$dF_{Iz} = dF_I \cdot \cos \theta = \left[P_o + \frac{2(\sigma_o + Kz)}{R} \right] \cdot (2\pi R dz) \cdot (-z/R)$$

$$F_{Iz} = -2\pi \int_{-R}^R \left[P_o + \frac{2(\sigma_o + Kz)}{R} \right] z dz$$

$$= -2\pi \int_{-R}^R \left(\frac{2Kz}{R} \right) z dz$$

$$= -\frac{8}{3} \pi R^2 K \dots\dots\dots(4)$$

Kaptay *et al.*²⁾ considered the interfacial force acting on the particle along x direction as the derivative of the total interfacial energy by x . But the work of the movement of a particle along x direction must be equal to the change of total energy of system. When a particle moves in the liquid with interfacial tension gradient, not only the total interfacial energy between the particle and liquid, but other kinds of energy such as the enthalpy (PV , P : pressure, V : volume) of liquid and particle also change. In addition, the movement of liquid and the viscous resistance will also result in the change of total energy of system. The change of total interfacial energy between particle and liquid is not equal to the change of total energy of system. Therefore, the interfacial force acting on the particle along x direction can not be derived from the derivative of the total interfacial energy.

Mukai and Lin³⁾ derived out the interfacial force as the particle is in the stationary state. It is unnecessary to consider the change of total energy of system as particle moves. In addition, they obtained the same description of F_I (Eq. (4)) as Eq. (3) by a method similar to that used for deriving buoyancy from stationary pressure acting on the particle that is located in fluid. Therefore, we think Eq. (3) derived by Mukai and Lin can describe the motion of fine bubbles and solid particles under the interfacial tension gradient.

Kaptay *et al.*²⁾ derived a larger interfacial force than that it should be. It can be demonstrated as follows.

Mukai and Lin³⁾ derived Eq. (3) by integrating $dF_{Iz} = dF_I \sin \theta = 2\pi y(\sigma_{PL1} - \sigma_{PL2}) \sin \theta = -2\pi y K dx \sin \theta$ from $x =$

Fig. 1. System under microgravity and interfacial tension gradient. P_o : atmospheric pressure, P_l : liquid pressure, P_p : pressure in particle.

$-R$ to R . Here σ_{PL1} and σ_{PL2} are interfacial tensions between particle and liquid 1, and between particle and liquid 2, respectively (Fig. 2 and Eq. (6) to Eq. (14) in reference 3). Equation (2) derived by Kaptay *et al.*²⁾ can be also obtained by integrating dF_{Lz} , $dF_{Lz,z=0} = 2\pi R(\sigma_{PL1} - \sigma_{PL2}) = -2\pi RKdz$ from $z = -R$ to R . $dF_{Lz,z=0}$ is the driving force acting on the infinite small volume AA'B'B located at the central line of the particle, as shown in Fig. 2.

$$dF_1 = dF_{Lz,z=0} = 2\pi R(\sigma_{PL1} - \sigma_{PL2}) = -2\pi RKdz \quad \dots\dots(5)$$

$$F_1 = -2\pi RK \int_{-R}^R dz = -4\pi R^2 K \quad \dots\dots\dots(6)$$

However, dF_{Lz} acting on the infinite small volume located at any another position of the particle except for $z=0$ is smaller than $dF_{Lz,z=0}$. Then F_{Lz} must be smaller than $4\pi R^2 K$. Obviously Eq. (2) derived by Kaptay *et al.*²⁾ overestimates the interfacial force.

3. The Flow Pattern of Liquid along the Surface Bubble

Kaptay *et al.*²⁾ indicated in their paper that a wake must

Fig. 2. Coordinate system for analyzing the driving force due to interfacial tension gradient.

form behind a sphere even if the Reynolds number, N_{Re} , is less than 1, and the separation point is located on the half of the sphere according to the reference 5⁴⁾ they quoted. But that reference⁴⁾ just said a wake might form behind the sphere that is located in both laminar and turbulent flow and the location of separation point was determined according to whether the flow is laminar or turbulent, while it did not say whether the wake can form in laminar flow with a N_{Re} less than 1. Furthermore, the reference⁴⁾ indicated that Stokes' law applies up to $N_{Re} \approx 1$ for spheres, which means no wake forms in this case because Stokes' law is not valid if wake forms.

Many other researchers⁵⁻⁷⁾ indicated that no wake could occur behind the moving spheroid if its N_{Re} is small. Szekely⁵⁾ indicated that the drag force acting on a sphere moving in fluid could be expressed with Stokes' law when N_{Re} is less than 2, which means no any other drag forces existed except the viscosity of fluid. In other word, no wake but laminar flow could occur around whole surface of sphere. Taneda⁶⁾ observed the flow pattern around the surface of a sphere at various Reynolds numbers (Fig. 3) and indicated that the critical Reynolds number at which the permanent wake begins to form in the rear of a sphere is about 24, *i.e.* the wake can not occur when N_{Re} is less than 24. Even if a wake occurs behind the sphere when the Reynolds number is large, the location of separation point (S in Fig. 3) changes with N_{Re} but not just located at the half of sphere as that Kaptay *et al.*²⁾ indicated in their paper.

Under the condition of our experiment, Reynolds number of bubble varied between 0.0056–0.17. So the liquid flow around the bubble should be that shown in Fig. 4, *i.e.* no wake could occur. The concentration gradient of surfactant is not disturbed and keeps constant along whole surface of bubble rather than only along half of the bubble, and so does the surface tension gradient.

4. Comparison of Theoretical and Experimental Velocities of Bubbles

Before making the comparison of theoretical and experi-

Fig. 3. Photos showing flow pattern in the wake immediately behind a sphere at different Reynolds number.⁶⁾

Fig. 4. Flow pattern of liquid and surface tension gradient around the surface of fine bubble.
($d\sigma/dx=k$ while $0 \leq X_S \leq 2R$)

Table 1. Experimental errors.

	R_B	k_0	$K_C^{(1)*}$	$D_L^{(1)}$
	10^{-6} m		$\text{mNm}^{-1}(\text{mol/m}^3)^{-1}$	10^{-9} m s $^{-2}$
Value		0.332	3.06	1.5
Error	$\pm 6\%$	± 0.02	± 0.26	± 0.35

* $C_0 = 0.37 \text{ mol/m}^3$

mental results, let's check the experimental errors that might occur in our research.¹⁾ They include the errors in measuring the size and velocity of bubble, and the parameters used for calculation such as partition coefficient, k_0 , concentration dependency of surface tension of surfactant water solution, K_C , and diffusion coefficient, D_L .

4.1. Errors in Measuring the Size of Bubble

Due to the resolution limit of photograph taken by high-speed video camera, error for measuring radius of bubble, R_B , is about $\pm 6\%$ because the periphery of bubble could not be seen clearly.

4.2. Errors in Measuring k_0 , D_L and K_C

Because of the limitation of experimental apparatus, experimental errors were also introduced in measuring the parameters used for calculation such as k_0 , D_L and K_C .

All experimental errors are shown in Table 1.

4.3. Comparison of Theoretical and Experimental Results

According to Sec. 3, the condition $X_S = 2R$ rather than $X_S = R$ is valid when Reynolds number is less than 1. Therefore Eq. (2) is valid in the case of our experiment if the opinion of Kaptay *et al.*²⁾ is correct. Then we can obtain Eq. (7) from Eq. (2) and Eq. (3).

$$V_{KK} = \frac{4}{8/3} V_{ML} = \frac{3}{2} V_{ML} \dots\dots\dots(7)$$

where "KK" and "ML" in the subscript of the velocity of the bubbles mean those derived from Eqs. (2) and (3), respectively.

Let's compare the measured velocity with calculated ve-

Fig. 5. Influence of C_0 on the velocity of the bubble.

locities by both Mukai *et al.*³⁾ and Kaptay *et al.*²⁾ opinions by taking into account the experimental errors that are shown in Table 1. The results are shown in Fig. 5. Areas with horizontal and vertical stripes are the velocity calculated from Eq. (7) and the velocity calculated from Eq. (5) of our paper¹⁾ by taking into account possible experimental errors, respectively.

According to Fig. 5, experimental result is within the range of calculated results from the equation derived by Mukai and Lin and is closed to the lower calculated results, while it is less than calculated results from Eq. (7) even considering the experimental errors.

In conclusion, we do not agree with Kaptay *et al.*'s opinion²⁾ in the following points: i) derivation process of Eq. (2) and ii) the wake formation in the range of $N_{Re} < 1$. We also point out that the comparison between calculated velocity and measured velocity cannot prove the validity of Kaptay *et al.*'s²⁾ opinion.

Acknowledgements

We appreciate Kaptay and Kelemen very much for their sincere concern to our researches. We believe that this discussion is very helpful for further clarifying the mechanism of surface tension gradient influencing the behavior of particles. We are also waiting for a new and valuable comment from Kaptay and Kelemen.

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