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ABSTRACT

Mass transfer from single carbon dioxide bubbles in vertical pipes is measured using a stereoscopic image processing method to develop a mass transfer correlation applicable to a wide range of bubble diameters in standard pipe sizes. The diameters of pipes used are 12.5, 18.2, and 25.0 mm and the bubble diameter ranges from 5 to 26 mm. The ratio, λ , of the bubble diameter to the pipe diameter is varied from 0.2 to 1.8, which covers various bubble shapes such as spheroidal, wobbling, cap, and Taylor bubbles. Measured Sherwood numbers, *Sh*, strongly depend on bubble shape, i.e., *Sh* of Taylor bubbles differs from that of spheroidal and wobbling bubbles. The Sherwood numbers are correlated in terms of the Peclet number *Pe* and λ . The applicability of the proposed correlation to long-term bubble dissolution process is examined through comparisons between measured and predicted dissolution processes. The predictions are carried out by solving mass conservation equations not only for carbon dioxide but also for nitrogen and oxygen. Good agreements are obtained in the dissolution processes for various bubble sizes and the three pipe diameters.

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1. Introduction

Mass transfer between bubbles and liquid has been utilized in various process engineering systems such as bubble columns, bioreactors, gas-to-liquid plants, sequestration of carbon dioxide (CO_2) in ocean [1] and so on. The mass transfer is known to depend on various factors such as bubble diameter, bubble shape, bubble velocity, fluid properties of the two phases, degree of liquid contamination, presence of a pipe wall and so on. A number of studies have, therefore, been carried out to understand the effects of these factors on the mass transfer [2–10].

Several models of the Sherwood number, *Sh*, for a single bubble rising through an infinite stagnant liquid have been proposed for various bubble diameters and for various bubble shapes such as spherical, spheroidal, wobbling, and cap bubbles. Boussinesq [2] theoretically investigated mass transfer from single spherical bubbles in a potential flow and proposed the following model:

$$Sh = \frac{2}{\sqrt{\pi}} P e^{1/2} \tag{1}$$

where Pe is the Peclet number defined by

$$Pe = \frac{V_B d}{D_C} \tag{2}$$

where V_B is the terminal velocity of a bubble, d the sphere-volume equivalent bubble diameter and D_c the diffusion coefficient of gas species in the liquid phase. Lochiel and Calderbank [3] deduced the following *Sh* model based on a boundary layer theory not only for spherical bubbles but also for single oblate-spheroidal bubbles in infinite stagnant liquids:

$$Sh = \frac{2}{\sqrt{\pi}} \left[1 - \frac{2.9}{Re^{1/2}} \right]^{1/2} Pe^{1/2} \quad \text{for a shperical bubble}$$
(3)

$$Sh = \frac{2}{\sqrt{\pi}} \left[\frac{2(1-E^2)^{3/2}}{3E(\sin^{-1}(1-E^2)^{1/2} - E(1-E^2)^{1/2})} \right]^{1/2} Pe^{1/2}$$

for a shpereroidal bubble (4)

Here the Reynolds number Re is defined by

$$Re = \frac{\rho_L V_B d}{\mu_l} \tag{5}$$

where ρ_L is the liquid density, μ_L is the liquid viscosity. *E* is the aspect ratio, which is the ratio of the maximum vertical dimension of a bubble to the maximum horizontal dimension. Eq. (3) reduces to Eq. (1) as $Re \rightarrow \infty$. Eq. (4) indicates that *Sh* of a spheroidal bubble depends on *Pe* and bubble shape. Johnson et al. [4] carried out experiments on mass transfer from single wobbling and cap bubbles for a wide range of *d* and *Re* (6 < *d* < 40 mm, 500 < *Re* < 20000) and proposed an empirical correlation. *Sh* correlations taking into account the effects of liquid contamination on mass transfer from spherical and oblate-spheroidal bubbles have been also proposed [5–7]. In

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contrast to the mass transfer from free rising bubbles, our knowledge on mass transfer from bubbles rising through a stagnant liquid in a vertical pipe is still insufficient.

Several studies have been carried out for mass transfer from single Taylor bubbles in a vertical pipe. Filla [8] carried out experiments on single CO_2 Taylor bubbles and proposed:

$$Sh_D = 5.1(L_B/D)^{0.8} P e_D^{0.5}$$
(6)

where L_B is the length of a Taylor bubble, *D* is the pipe diameter, and Sh_D and Pe_D are the Sherwood number and the Peclet number defined by using *D* as the characteristic length:

$$Sh_D = \frac{k_L A_T}{D_C D} \tag{7}$$

$$Pe_D = \frac{V_B D}{D_C} \tag{8}$$

where k_L is the mass transfer coefficient and A_T the surface area of a Taylor bubble. The applicable range of Eq. (6) is $1 < L_B/D < 6$. Eq. (6) requires an additional model for L_B/D . As for the effects of the pipe wall on mass transfer from small bubbles (spheroidal, wobbling, and cap bubbles), Clift et al. [9] speculated that the ratio λ of d to D does not affect the mass transfer when λ is less than 0.5. Abe et al. [10] carried out experiments on mass transfer from CO₂ bubbles for a wide range of λ , i.e., $0.2 < \lambda < 1.0$, which covers not only small bubbles but also Taylor bubbles. They proposed the following correlation:

$$Sh = 0.0818 \text{Re}^{0.372} P e^{0.5} \tag{9}$$

which is based on their experimental data obtained by using a pipe of D = 25 mm. Hence the applicability of Eq. (9) to different pipe diameters is not clear. Tsuchiya et al. [11] modified the Clift's *Sh* correlation [9] by taking into account the surface renewal effect, and applied it to the prediction of long-term bubble dissolution. However, the predictions were not in good agreement with their experimental data.

Mass transfer from single CO_2 bubbles in vertical pipes is, therefore, measured in this study to develop a *Sh* correlation applicable to various bubbles in a vertical pipe. The pipe diameters used are 12.5, 18.2, and 25.0 mm to examine effects of λ on *Sh*. A numerical prediction of bubble dissolution is also carried out to examine the applicability of the proposed *Sh* correlation to long-term dissolution process.

2. Experiments

2.1. Experimental setup

Fig. 1 shows a schematic of the experimental apparatus, which consists of the test section, the lower and upper tanks, the two high-speed video cameras, the two LED light sources, the two optical filters, the four *z*-axis stage actuators and the digital fiber sensor. The test section was a vertical pipe of 2000 mm long. Three pipes, 12.5, 18.2, and 25.0 mm in inner diameter, were used. The reference elevation (z = 0 mm) in measurements was located 1900 mm below the water surface in the upper tank. The pipe was made of fluorinated-ethylene-propylene (FEP) resin, whose refractive index is close to that of water, i.e., the refractive indexes of FEP resin and water are 1.338 and 1.333, respectively. The FEP pipe was installed in the acrylic duct and water was filled in the gap between the duct and the pipe, which enabled observation of bubbles without any optical distortion.

A predetermined amount of CO_2 gas (99.9 vol.% purity), whose volume was measured by using the gastight syringe, was stored in the hemispherical cup. A single bubble was released by rotating



Fig. 1. Experimental setup.

the cup. Water purified by using a Millipore system (Elix 3) was used for the liquid phase. The temperature T was fixed at 298 ± 1 K.

As shown in Fig. 2, front and side images of a bubble were recorded by using the two synchronized video cameras (frame rate: 250 frame/s, exposure time: 1000 µs, spatial resolution: 0.04 mm/ pixel) mounted on the two actuators, which were placed at 90 degree around the pipe. The two color LED lights and optical filters reduced overexposure caused by light reflection at bubble surface. The motions of the cameras and the LED lights were synchronized using the actuators was adjusted at a constant value close to the bubble rising velocity. The digital fiber sensor detected the arrival of a bubble at the elevation of the relay control circuit. Thereby, the heights of the lights, bubble, and cameras were automatically synchronized. This stereoscopic bubble tracking system enabled us to record high-resolution bubble images.

When measuring long-term bubble dissolution processes, single bubbles were kept almost stationary at $z \approx 0$ mm in downward flow until the mass transfer reached an equilibrium condition.

An image processing method [10,12] was used to measure instantaneous bubble volumes, diameters, and positions. An example of a set of original bubble images is shown in Fig. 3(a). The front



Fig. 2. Optical setup.

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