Dissolution of a Carbon Dioxide Bubble in a Vertical Pipe*

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Abstract
Dissolution of single carbon dioxide (CO\(_2\)) bubbles in a vertical pipe of 25 mm diameter is measured to examine the effects of the ratio \(\lambda\) of the sphere–volume equivalent bubble diameter to the pipe diameter, the liquid Reynolds number and surfactants on mass transfer. The bubble diameter and liquid Reynolds number are varied from 5.0 to 26 mm (0.20 < \(\lambda\) < 1.0) and from 0 to 3100, respectively. Millipore water, tap water or water contaminated with Triton X–100 are used for the liquid phase. Dissolution processes are measured at atmospheric pressure and room temperature. Mass transfer coefficients and Sherwood numbers are evaluated from measured bubble diameters. Complicated capillary waves are formed at the clean bubble surface, whereas there are no capillary waves at the contaminated bubble surface. The disappearance of capillary wave results in the retardation of surface renewal, and therefore, Sherwood number decreases with increasing surfactant concentration. Empirical correlations of Sherwood numbers for bubbles rising in clean and contaminated liquids in a vertical pipe are proposed. The correlations are applicable not only to bubbles in stagnant liquid but also to bubbles in pipe flow, provided that the liquid Reynolds number is not so high.

Key words: Mass Transfer; Bubble Dissolution; Carbon Dioxide (CO\(_2\)); Surfactant

1. Introduction

Mass transfer between bubbles and the surrounding liquid has been utilized in various process engineering systems such as bioreactors, gas–to–liquid (GTL) plants, melting and refining processes of glass and steel, carbon dioxide (CO\(_2\)) sequestration in ocean (1). The knowledge of mass transfer is indispensable for improving the quality of products, reducing the costs in industrial processes, evaluating the feasibility of the sequestration system and so on.

Mass transfer from a single bubble depends on various factors such as fluid properties of the two phases, bubble size, bubble shape, bubble rising velocity and surfactant concentration. A number of studies on single bubbles dissolving in infinite stagnant liquid have, therefore, been conducted, and many models and empirical correlations for the mass transfer coefficient \(k_L\) and the Sherwood number \(Sh\) have been proposed (2)–(4). However, little is known on mass transfer from a bubble flowing in a pipe flow. Clift et al. (5) discussed the influence of the diameter ratio \(\lambda\) (= \(d\) / \(D\), where \(d\) is the sphere–volume equivalent bubble diameter and \(D\) is the pipe diameter) on mass transfer from single bubbles in stagnant liquid filled in a vertical pipe. They pointed out that mass transfer from a bubble of \(\lambda\) < 0.5 is not so much different from that from a bubble in infinite stagnant liquid.

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liquid. For Taylor bubbles ($\lambda > 1.0$) in stagnant liquid, Filla (6) and Niranjan et al. (7) proposed empirical correlations of $k_L$. However we lack the knowledge on single bubbles of 0.5 < $\lambda$ < 1.0. In addition, there are no available correlations of $k_L$ and $Sh$ for single bubbles rising in a liquid flow in a vertical pipe, since no experimental data on dissolving bubbles in a pipe flow have been reported.

Many studies (8)–(11) have pointed out that mass transfer depends on the degree of contamination of bubble surface because surfactants adsorbed at bubble surface retard mass transfer from a bubble. However, few studies have dealt with mass transfer from a large contaminated bubble in a pipe.

The effects of the diameter ratio, bulk liquid flow and surfactants on dissolution of a distorted CO$_2$ bubble in a vertical pipe are, therefore, experimentally investigated in this study.

Nomenclature

$A$ Surface area of volume equivalent sphere = $\pi d^2$, m$^2$

$C$ Concentration of CO$_2$ in water, mol m$^{-3}$

$C_L$ Concentration of H$_2$O in water, mol m$^{-3}$

$C_S$ Concentration of CO$_2$ at gas–liquid interface, mol m$^{-3}$

$C_{surf}$ Concentration of surfactant in contaminated liquid, mol m$^{-3}$

$d$ Sphere–volume equivalent bubble diameter, m

$D$ Pipe inner diameter, m

$D_C$ Diffusion coefficient of gas in liquid, m$^2$ s$^{-1}$

$g$ Acceleration of gravity, m s$^{-2}$

$H$ Henry’s constant, Pa

$k_L$ Liquid–phase mass transfer coefficient of volume equivalent sphere, m s$^{-1}$

$n$ Total moles of gas inside a bubble, mol

$P$ Pressure inside a bubble, Pa

$R$ Universal gas constant, J K$^{-1}$mol$^{-1}$

$Re$ Bubble Reynolds number = $\rho_L V_R d / \mu_L$

$Re_L$ Liquid Reynolds number = $\rho_L \overline{V_L} D / \mu_L$

$Sc$ Schmidt number = $\mu_L / \rho_L D_C$

$Sh$ Sherwood number = $k_L d / D_C$

$t$ Time, s

$T$ Temperature, K

$V_R$ Rising velocity of a bubble, m s$^{-1}$

$\overline{V_L}$ Mean liquid velocity, m s$^{-1}$

$V_R$ Relative velocity between a bubble and liquid, m s$^{-1}$

$V_T$ Terminal rising velocity of a bubble, m s$^{-1}$

$z$ Vertical position, m

Greek symbols

$\Delta l_p$ Height of the elliptical disk equivalent to one pixel of a bubble image, m

$\lambda$ Ratio of sphere–volume equivalent diameter to pipe inner diameter, $d / D$

$\mu_L$ Liquid viscosity, Pa s

$\rho_L$ Liquid density, kg m$^{-3}$

$\sigma$ Surface tension, N m$^{-1}$

2. Experimental

Figure 1 shows a schematic of the experimental apparatus, which consists of the test section, the lower tank, the upper tank, the pump, the flowmeter, two high–speed video
cameras, two LED light sources, two optical filters, four z-axis stage actuators and the digital fiber sensor. The test section was the vertical pipe of 25 mm diameter $D$ and 2000 mm long. The pipe was made of fluorinated-ethylene-propylene (FEP) resin, whose refractive index is very close to that of water. The refractive indexes of FEP resin and water are 1.338 and 1.333, respectively. The FEP pipe was installed in the acrylic duct. Water was filled in the space between the duct and the pipe. Hence, we could observe bubbles without any optical distortion.

A predetermined amount of CO$_2$ (99.9 vol.% purity), the volume of which was measured by using the gastight syringe, was stored in the hemispherical cup. A single bubble was released by rotating the cup. The dissolution processes of single CO$_2$ bubbles ranging from 5.0 to 26 mm in $d$ ($\lambda = 0.20 - 1.0$) were measured at atmospheric pressure and room temperature. Millipore water, tap water or water contaminated with a surfactant, Triton X–100 (Wako Pure Chemical Industries, 168–11805) were used for the liquid phase. A Millipore system (Elix 3) was used to purify tap water. The concentration $C_{surf}$ of Triton X–100 in Millipore water was 0.50, 1.0 or 10 mmol/m$^3$. Single bubbles in tap water were measured under three liquid Reynolds number conditions, $Re_L = 0$, 1600 and 3100 to examine the effects of $Re_L$ on mass transfer. Here, $Re_L$ is defined by

$$Re_L = \frac{\rho_L \bar{V}_L D}{\mu_L}$$

(1)

where the subscript $L$ denotes the liquid phase, $\rho$ the density, $\bar{V}_L$ the mean velocity ($\bar{V}_L = 0, 0.057$ and 0.11 m/s), and $\mu$ is the viscosity. Water temperature was kept at 298 + 1.0 K, at which $\rho_L$ and $\mu_L$ are 997 kg/m$^3$ and 0.890 x 10$^{-3}$ Pa s, respectively.

As shown in Fig. 2, front and side images of a bubble were recorded by using the two synchronized video cameras (Kodak Motion Corder Analyzer SR–500, frame rate: 250 frame/s, exposure time: 1.0 ms), which were mounted on the two actuators (SUS Corp.)

![Fig. 1 Schematic of the experimental apparatus.](image)
The green and red LED light sources (HAYASHI WATCH–WORKS, AHL–3000–05G0: green, HDB90x90R: red) were on the other actuators. The two color LED lights and optical filters reduced the overexposure of images, i.e., the so–called blown out, caused by the reflection of light at bubble surface. All the actuators were synchronized with the video cameras by a relay control circuit. The translational speed of the actuators was adjusted to a constant value close to the bubble rising velocity. The digital fiber sensor (OMRON, E32–T16WR) detected the arrival of a bubble below the actuators. The amplifier (OMRON, E3X–DA11–S) subsequently triggered the relay control circuit. Thereby, the heights of the light source, a rising bubble and the camera were automatically synchronized. This optical arrangement enabled us to record clear bubble images. The spatial resolution of images was 0.10 mm/pixel.

An image processing method proposed in our previous studies (12)(13) was utilized to measure bubble volumes, bubble diameters and bubble positions. An example of original images of a bubble is shown in Fig. 3 (a). The front and side images were transformed into binary images (Fig. 3 (b)). The method assumes that all the horizontal cross–sections of a bubble are elliptical. The height of each elliptic disk in the image is one pixel and its physical length is $\Delta l_p$. The lengths of the major and minor axes of the elliptic disk were obtained from the lengths of front and side images in the horizontal plane ($R_i$ and $G_i$, Fig. 3 (c)). The resultant elliptic disks were piled up in the vertical direction to reconstruct a three–dimensional bubble shape as shown in Fig. 3 (d).

The sphere–volume equivalent bubble diameter $d$ is evaluated by

$$\frac{\pi d^3}{6} = \sum_{i=1}^{N} \left( \frac{\pi R_i G_i \Delta l_p}{4} \right)$$

(2)

where $N$ is the total number of disks in the vertical direction of the bubble image.

Instantaneous bubble volumes, diameters and positions were evaluated from the reconstructed bubble shapes. Bubble rising velocities $V_B$ were evaluated from the axial position $z$ of a bubble. Here, the origin of $z$ was defined at 1.85 m below the free surface as shown in Fig. 1. Measurement uncertainties estimated at 95 % confidence in $d$ and $V_B$ were $\pm 2.1 \%$ and $\pm 1.0 \%$, respectively.

Mass transfer coefficients $k_L$ and Sherwood numbers $Sh$ were evaluated from the rate of decrease in bubble diameter. Assuming that flow is isothermal and CO2 concentration in a bubble is 100 %, the rate of mass transfer from a bubble to water, $dn/dt$, is expressed as

$$\frac{dn}{dt} = k_L A(C_s - C)$$

(3)
where $n$ is the total moles of CO$_2$ in a bubble, $t$ the time, $A (= \pi d^2)$ the bubble surface area, $C_S$ the CO$_2$ concentration at gas–liquid interface, and $C$ the CO$_2$ concentration in water.

Assuming that $C_S$ is determined by the Henry’s law and $C$ is negligible, we obtain

$$k_L = \frac{1}{\pi d^2} \frac{H - P(z)}{C_i P(z)} \frac{dn}{dt}$$  \hspace{1cm} (4)

where $H$ is the Henry’s constant (166 MPa for CO$_2$ at $T = 298$ K), $C_L$ the H$_2$O concentration (55.4 kmol/m$^3$) and $P(z)$ the pressure inside a bubble given by

$$P(z) = P_{atm} + \rho_L g (1.85 - z) + \frac{4\sigma}{d}$$  \hspace{1cm} (5)

where $P_{atm}$ is the atmospheric pressure, $g$ the acceleration of gravity and $\sigma$ the surface tension. By assuming that CO$_2$ is an ideal gas, $dn/dt$ is expressed in terms of $P(z)$ and $d$ as follows:

$$\frac{dn}{dt} = \frac{\pi}{6RT} \frac{d(d^3 P(z))}{dt}$$  \hspace{1cm} (6)

where $R$ is the universal gas constant and $T$ the temperature. Substituting Eq. (6) into Eq. (4), and evaluating $d(d^3 P(z))/dt$ by using measured pressures and bubble diameters at two positions $z_1$ and $z_2$, we obtain

$$k_L = \frac{(H - P(z))(d_{z_1}^{-3} P(z_1) - d_{z_2}^{-3} P(z_2))}{6RT \Delta d^2 C_i P(z)}$$  \hspace{1cm} (7)
where $\Delta t$ is the time duration of bubble translation from $z_1$ to $z_2$. The above equation is used to evaluate $k_L$ and the Sherwood number:

$$Sh = \frac{k_L d}{D_C}$$

(8)

where $D_C$ is the diffusion coefficient, which is $1.9 \times 10^{-9}$ m$^2$/s for CO$_2$ in water at $T = 298$ K and atmospheric pressure$^{14}$.

3. Results and Discussion

3.1 Effects of diameter ratio

The mass transfer coefficient $k_L$ for single bubbles in clean stagnant water filled in the pipe is plotted against the diameter ratio $\lambda$ in Fig. 4. The large scatter in $k_L$ is caused by a small error in measured $d$. In spite of the increase in $d$, $k_L$ decreases with increasing $\lambda$. Johnson et al. $^{3}$ proposed the following empirical correlation of $k_L$ for single distorted bubbles of $6 < d < 40$ mm in infinite stagnant water:

$$k_L = \frac{2}{\sqrt{\pi}} \left( \frac{d}{0.0045 + 0.2d} \right)^{1/2} \left( \frac{V_R D_C}{d} \right)^{1/2} \text{ for } 500 < Re < 20000$$

(9)

$$Re = \frac{\rho_l V_R d}{\mu_l}$$

(10)

where $V_R$ is the relative velocity between a bubble and the liquid and $Re$ the bubble Reynolds number. The broken curve in Fig. 4 is drawn using the combination of Eq. (9) and an empirical correlation of $V_R$ for single bubbles in a vertical pipe proposed by Nakahara & Tomiyama$^{15}$. The detail on the $V_R$ correlation is given in Appendix. Johnson’s correlation agrees well with the measured $k_L$, which, in turn, implies that a mass transfer model developed for a single bubble in infinite stagnant liquid is applicable to a single bubble in a vertical pipe, provided that a correct relative velocity between a bubble and the liquid is substituted into the mass transfer correlation.

![Fig. 4 Mass transfer coefficient of a bubble in stagnant Millipore water.](Image)
Since Eq. (9) includes the term with dimension, $d$, the following empirical correlation, which consists of only dimensionless groups, was derived from the experimental data:

$$Sh = 0.0818Re^{0.472}Sc^{1/2}$$  \hspace{1cm} (11)

![Fig. 5](image1)

**Fig. 5**  Sherwood number of a bubble in stagnant Millipore water.

![Fig. 6](image2)

**Fig. 6**  Sherwood number of a bubble in pipe flows.
\[ Sc = \frac{\mu_\lambda}{\rho_i D_e} \]  

(12)

where \( Sc \) is the Schmidt number. As shown in Fig. 5, both Eq. (9) and Eq. (11) can be used to evaluate the mass transfer from a single bubble rising through clean stagnant liquid in a vertical pipe for \( 0.20 < \lambda < 1.0 \) and \( 1100 < Re < 4700 \).

### 3.2 Effects of Liquid Reynolds number

Sherwood numbers measured for \( Re_L = 0 \), 1600 and 3100 are plotted against \( Re \) in Fig. 6. The Sherwood number does not depend on \( Re_L \). That is, mass transfer from single CO\(_2\) bubbles is independent of the bulk liquid flow. This might be because the magnitude of turbulent fluctuation velocity \( u' \) is much smaller than the relative velocity \( V_R \) (\( u' \) is about 0.01 m/s for \( Re_L = 3100 \) and \( V_R \) ranges from 0.15 to 0.25 m/s). Since there are no effects of bulk liquid flow on mass transfer, we can apply Eq. (11) to single bubbles in pipe flow as shown in Fig. 6.

### 3.3 Effects of surfactants

Velocities and mass transfer coefficients of single bubbles rising through stagnant water filled in the pipe were measured using Millipore water, tap water, or water contaminated with Triton X–100 (\( C_{surf} = 0.50 \), 1.0 and 10 mmol/m\(^3\)). As shown in Fig. 7, \( V_B \) does not depend on the quality of water, which might be because the bubble diameters are large enough to be mainly governed by the inertial force\(^{16}\).

As shown in Fig. 8, there is no difference in \( Sh \) between tap water and Millipore water, and the Sherwood numbers for these two water systems are well predicted by Eq. (11). The increase in surfactant concentration causes a large decrease in \( Sh \). The experimental data for \( C_{surf} = 10 \) mmol/m\(^3\), i.e., \( Sh \) for fully–contaminated bubbles is well expressed by the following correlation:

\[ Sh = 7.52 \times 10^{-5} Re^{1.76} Sc^{1/3} \]  

(13)

Figure 9 shows sequential images of single CO\(_2\) bubbles rising in Millipore water and in the contaminated water (\( C_{surf} = 1.0 \) mmol/m\(^3\)). Complicated capillary waves are formed at the clean bubble surface, whereas there are no capillary waves at the contaminated bubble.
surface. This must be because the damping coefficient of capillary wave becomes much higher when surfactants are accumulated at bubble surface $^{(12)(17)}$. The disappearance of

Fig. 8 Sherwood number of a bubble in stagnant waters.

Fig. 9 Sequential images of a bubble ($d = 9$ mm) in Millipore water and in contaminated water.
capillary wave results in the retardation of surface renewal\(^{(18)}\), and therefore, \(Sh\) decreases with increasing surfactant concentration.

4. Conclusion

Dissolution of single carbon dioxide (CO\(_2\)) bubbles rising in a vertical pipe of 25 mm diameter was measured to understand the effects of the ratio \(\lambda\) of the bubble diameter to the pipe diameter, the liquid Reynolds number \(Re_L\) and surfactants on mass transfer. The bubble diameter and liquid Reynolds number ranged from 5.0 to 26 mm (0.20 < \(\lambda\) < 1.0) and from 0 to 3100, respectively. Clean water, tap water and water contaminated with Triton X–100 were used for the liquid phase. Mass transfer coefficients and Sherwood numbers were evaluated from measured bubble diameters. As a result, the following conclusions were obtained.

(a) The mass transfer coefficient for bubbles in stagnant clean water decreases with increasing the diameter ratio \(\lambda\).

(b) The Sherwood number does not depend on \(Re_L\) because the magnitude of turbulent fluctuation velocity is much smaller than a relative velocity.

(c) Empirical correlations of Sherwood numbers for bubbles rising in clean and contaminated liquids in a vertical pipe are proposed. The correlations are applicable not only to bubbles in stagnant liquid but also to bubbles in pipe flow, provided that the liquid Reynolds number is not so high.

(d) Mass transfer coefficients for bubbles in contaminated water with Triton X–100 become smaller than those in clean water owing to the disappearance of capillary wave.

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Appendix

Nakahara & Tomiyama\(^{(15)}\) proposed the following correlations of \(V_T\) for a single bubble rising in a stagnant liquid in a vertical pipe under the condition of 0.2 < \(\lambda\) < 2.0 and 13 < \(D\) < 25 mm.

\[
V_T = \begin{cases} 
V_{T0} \phi(\lambda)^{-1/2} & \lambda \leq 0.6 \\
Fr \left[ \left( \frac{\rho_L - \rho_G}{\rho_L} \right) D \right]^{1/2} & 0.6 < \lambda
\end{cases}
\]  \(\text{(A1)}\)

where \(V_{T0}\) is the terminal velocity in an infinite stagnant liquid\(^{(12)}\), \(Fr\) the Froude number and \(\phi(\lambda)\) is the wall effect multiplier given by

\[
\phi(\lambda) = \begin{cases} 
1, & 0 \leq \lambda \leq \lambda^* \\
\frac{1}{4} \left[ 1.13 \exp(-\lambda) + (1-\lambda^2)^{3/2} \right]^{2}, & \lambda^* < \lambda \\
\frac{1}{4} \left[ 1.13 \exp(-\lambda^*) + (1-\lambda^2)^{3/2} \right]^{2}, & \lambda^* < \lambda
\end{cases}
\]  \(\text{(A2)}\)

\[
\lambda^* = 0.394 \exp(2.93 E_{OD}^{-0.5})
\]  \(\text{(A3)}\)

\[
E_{OD} = \frac{g(\rho_L - \rho_G) D^{1/2}}{\sigma}
\]  \(\text{(A4)}\)

where \(\lambda^*\) represents the boundary between the distorted ellipsoidal shape and Taylor bubble shape and \(E_{OD}\) the Eötvös number which is a function of \(D\). According to Wallis\(^{(19)}\), \(Fr\) is given by
\[ Fr = 0.345 \left[ 1 - \exp \left( \frac{3.37 - Eo_D}{10} \right) \right] \]  

(A5)

References