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## Effects of a bubble and the surrounding liquid motions on the instantaneous mass transfer across the gas–liquid interface

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## HIGHLIGHTS

- Elucidating mass transfer process of a zigzagging bubble based on precise experiments.
- A clear understanding of roles of bubble motions and the surrounding liquid motion.
- Mass transfer coefficients of the zigzagging CO<sub>2</sub> bubble in a time interval of 6 ms.
- The coefficients gained by a smart experimental technique and a new image processing.

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## ABSTRACT

To understand the mass transfer mechanism from a bubble to the surrounding liquid, one must consider the relation between the mass transfer, gravity-center and surface motions of the bubble. Bubbles in chemical and bio-reactors usually exhibit a zigzag motion. Simultaneously, such bubbles accompany periodic surface oscillation. Knowledge about the correlation between the mass-transfer mechanism and bubble motion is as yet incomplete. In this study, we used experimental results from highly precise measurements of bubble volume to clarify the instantaneous mass-transfer coefficients of a zigzagging CO<sub>2</sub> bubble. We visualized the zigzagging motion and surface oscillation 3-dimensionally and simultaneously, using two high-speed cameras and mirrors. We also visualized the CO<sub>2</sub> dissolution (mass transfer) process from the bubble to the surrounding liquid using the LIF/HPTS method. To obtain the precise instantaneous mass transfer coefficient and bubble motions, single bubbles were visualized in three sections: the linear-ascent, the first-inversion, and the second-inversion of the zigzag motion. The instantaneous mass-transfer coefficients in an interval of 6 ms in these sections were calculated from the bubble-volume shrinkage with ms time-resolution. The instantaneous mass-transfer coefficients increased in acceleration areas—i.e., in the linear-ascent and second-inversion sections. Interestingly, in the latter, the gravity-center velocity of the bubble reached terminal velocity and was constant, but the velocity of the bubble hemisphere was accelerated due to the zigzag motion. This partial acceleration of the bubble hemisphere led to a high renewal rate of the liquid on the bubble interface. The effect of the partial acceleration on the instantaneous mass transfer was significant for bubbles categorized into zigzag motion.

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## 1. Introduction

A deep knowledge of multiphase flows is essential for many industrial applications, such as bioreactors, chemical reactors and power plants. In particular, a bubbly flow is commonly encountered in industrial processes (e.g., chemical reaction, agitation, oxi-

dation, fermentation, and dissolution) for enhancing a mass transport rate and/or a chemical reaction rate. For instance, the Gas Lift Advanced Dissolution (GLAD) system, a promising option for mitigating global warming, has been proposed [1–4]. The GLAD system involves submerging an inverse-J pipeline in the ocean. CO<sub>2</sub> bubbles injected into the pipe from a power plant dissolve in seawater as they rise, and transport the CO<sub>2</sub>-rich seawater to a great depth. Thus the CO<sub>2</sub> can be sequestered with high efficiency at a great depth due to energy reduction in CO<sub>2</sub> transportation and

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## Nomenclature

$A_n$	Fourier cosine coefficient (mm)	$(r, \theta)$	polar coordinate system (mm, rad)
$B_n$	Fourier sine coefficient (mm)	$r$	length from bubble's center of gravity to contour (mm)
$c(x_c, z_c)$	center of gravity of the bubble (mm, mm)	$S$	surface area (mm <sup>2</sup> )
$c_S$	CO <sub>2</sub> concentration at the bubble interface (kg/m <sup>3</sup> )	$u_x$	horizontal velocity (mm/s)
$c_\infty$	CO <sub>2</sub> concentration of the bulk (kg/m <sup>3</sup> )	$u_z$	vertical velocity (mm/s)
$c_S^*$	average CO <sub>2</sub> concentration at the whole bubble interfacial area (kg/m <sup>3</sup> )	$U$	velocity of the bubble (mm/s)
$c_\infty^*$	average CO <sub>2</sub> concentration of the bulk (kg/m <sup>3</sup> )	$U_{\text{right}}$	velocity of the right-side edge of the bubble (mm/s)
$g_{b1}(x, z)$	bubble shape function of $x$ - $z$ plane (-)	$U_{\text{left}}$	velocity of the left-side edge of the bubble (mm/s)
$g_{b2}(y, z)$	bubble shape function of $y$ - $z$ plane (-)	$V_b(x, y, z)$	circumscribed cuboid function (-)
$g_{b3}(\zeta, \eta)$	bubble shape function of $\zeta$ - $\eta$ plane (-)	$(x_c, y_c)$	coordinates of bubble contour (mm, mm)
$I_1(x, z)$	bubble shape function of $x$ - $z$ plane (-)	$x$	coordinate (mm)
$k_L$	mass transfer coefficient (mm/s)	$y$	coordinate (mm)
$k_L^*$	surface average mass transfer coefficient (mm/s)	$z$	coordinate (mm)
$l_{\text{major}}$	major axis length of the bubble on $x$ - $z$ plane (mm)	$\Delta t$	time interval (s)
$l_{\text{minor}}$	minor axis length of the bubble on $y$ - $z$ plane (mm)	$\Delta V$	volume shrinkage in $\Delta t$ (mm <sup>3</sup> /s)
$N$	mass flux (kg/(m <sup>2</sup> s))	$\kappa_R$	curvature of the right edge of the bubble (-)
$N_{\text{ave}}$	average mass flux (kg/(m <sup>2</sup> s))	$\kappa_L$	curvature of the left edge of the bubble (-)
		$\rho$	density (kg/m <sup>3</sup> )

elimination of the need to purify and liquefy the CO<sub>2</sub>. To minimize the environmental impact of CO<sub>2</sub> sequestration on the deep ocean, the CO<sub>2</sub> dissolution and gas-lift effect must be controlled.

Since the early 1970s, researchers have been studying mass/heat transfer in bubbly flows, investigating average bubble diameters and average void fractions [5–9]. Bubbly flows are multiscale ranging phenomena and have typical nonlinearity; i.e., just like single-phase turbulent flows, their large-scale structures are intensively influenced by the boundary conditions [10]. The large-scale flow structure is essential for the enhancement of reaction and mixing in reactors, and is composed of smaller-scale flow structures. The small-scale flow structures are also essential to a deep understanding of the features in reacting and mixing processes. Bubbles in a bubbly flow show a wide variety of bubble size/velocity distributions, and their mutual interactions (breakup, coalescence, and bounce) are decisive factors in the large-scale flow structure of reactors.

Considerable research has considered the mass transfer from a single bubble, both experimentally and numerically [11–17]. It is well known that bubbles can be categorized based on size, and with regard to their shape and motion [18,19]. Mass transfer rates from a bubble to the surrounding liquid change according to its size and motion. The mass transfer from a large bubble, e.g., a spherical cap bubble, has been measured and discussed by Baird and Davidson [11] (in the range of 8–42 mm in diameter), Leonard and Houghton [12] (5–20 mm in diameter), and Calderbank and Lochiel [13] (4–31 mm in diameter). These researchers discussed the effects of bubble velocity, size, residence time, and the impurity of the liquid phase on the mass transfer of large bubbles. Takemura and Yabe investigated mass transfer from a small bubble, such as a spherical bubble (about 0.3 mm in diameter) [15,16]. They measured bubble size and ascent velocity from captured images of the bubble, and estimated the drag coefficient and Sherwood number, both experimentally and numerically. Moderate-sized bubbles (2–3 mm in diameter), e.g., oblate ellipsoidal bubbles, are known to show higher mass transfer coefficients than either larger or smaller bubbles [14].

Asymmetrical oblate ellipsoids with surface oscillation have been examined and investigated by many researchers [20–24]; consequently, much useful knowledge is available. Saffman [25] and Moore [26] were among the earliest to investigate zigzag and spiral bubble motion. In the past 20 years, a great deal of

information has been obtained on bubble-wake dynamics (e.g., horseshoe-like vortices) [27–29].

Bubble and wake motions affect mass transfer rates significantly in terms of the convective transportation. Although many previous researchers have investigated bubbles ranging from 2 to 3 mm in diameter, the enhancement mechanisms of the mass transfer coefficient are still uncertain due to the fluid dynamical complexity of these bubbles. In particular, interactions between the bubble motions (surface motion and centroid motion) and the surrounding liquid motion have yet to be elucidated.

In the present study, we precisely measured instantaneous mass transfer from the bubble to the surrounding liquid (mass transfer coefficient in 10-ms order short time interval). Simultaneously, we measured the bubble surface motion and centroid motion. Furthermore, we visualized the bubble wake motion. Focusing on the relationships among the instantaneous mass transfer coefficient, the bubble surface motion, centroid motion and the bubble wake, we attempted to clarify the enhancement mechanism of the mass transfer (i.e., the effects of the complex interactions on the mass transfer), based on highly precise experiments. For this specific purpose, we developed a new image processing code to measure very small volume changes in a zigzagging bubble. The bubble zigzag motion and surface oscillation were visualized 3-dimensionally (3D) and simultaneously, using two high-speed cameras and mirrors. In order to consider the effect of the characteristic liquid motion on the mass-transfer, we visualized dynamical bubble wake by the LIF/HPTS method (HPTS: 8-hydroxypyrene-1,3,6-trisulfonic acid, trisodium salt). Finally, we discuss major factors enhancing mass transfer from the bubble to the ambient liquid by comparing the instantaneous mass transfer, bubble motions and wake dynamics.

## 2. Experimental setup

Two types of experiments were carried out to accomplish the thorough analysis of the mass transfer phenomena from a zigzagging CO<sub>2</sub>-bubble to the surrounding liquid. To precisely measure the amount of CO<sub>2</sub> dissolution in the bubble, we captured bubble projection images using the experimental setup described below in Section 2.2. To analyze the dissolution process from the bubble, we employed the LIF/HPTS method described in Section 2.3, and

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