Numerical calculation of rising behavior and mass transfer for single CO\textsubscript{2} bubble in water

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Abstract. The rising behavior and mass transfer process of CO\textsubscript{2} bubble in the water were simulated and analyzed. The VOF model was applied to track the interface boundary. The CO\textsubscript{2} was added through an orifice with a diameter of 2.4 mm at the bottom. The bubble shapes through the rising process were shown and the aspect ratio was used to reflect the bubble deformation. The flow patterns around the bubble were also analyzed and its influence on mass transfer was discussed. The results show that the bubble equivalent diameter keeps constant though the bubble shapes change dramatically. Besides, the mass transfer process in the bubble rise section is different from that in the bubble formation and detachment sections.

1. Introduction

Bubble rising phenomenon often happens in the practical production processes, especially for distillation and reaction processes. The behaviors are complex when the conditions change. Bubble trajectories are quite different in various liquids and some parameters of bubbles, such as dimension, rising velocity, and flow patterns inside or outside the bubbles are also different. When the gas substances can be absorbed into the liquids, the mass transfer processes always occur with rising.

Mass transfer processes have been investigated by experiments in different devices and the results deepen the understandings of relevant fields. Hosoda et al. [1] studied the CO\textsubscript{2} bubble rose through water in a vertical pipe. Three kinds of pipes with different diameters were used and different initial bubbles behaviors were investigated. The ratio of pipe diameters and bubble diameters are applied as an important variable. In general, the rising velocity increases with bubble diameter increasing. However, the velocity stops increasing under some ratios in the vertical pipes. Besides, a diameter correlation was presented and its predictive accuracy was verified to be good. Aoki et al. [2] investigated the CO\textsubscript{2} bubble rising in the contaminated water based on the works of Hosoda et al. Only the pipe with a diameter of 12.5 mm was applied and the liquid was contaminated by surfactant. The mass transfer coefficient of contaminated water is lower compared with that of purified water and it decreases with the content of surfactant increasing. Then, Aoki et al. [3] studied the bubbles behaviors in alcohol aqueous solutions. Four different solutions, which are 1-pentanol, 1-heptanaol, 1-octanol, and 1-decanol, are applied as the liquids. The critical bubble diameters are reported under different
pipe diameters. As for mass transfer, the coefficient of bubbles decreases with content of alcohol solutions increasing.

The CO2 bubbles are more or less soluble in most liquids so that their diameters reduced continuously. Relevant experiment methods are quite important to improve the study accuracy. Nock et al. [4] analyzed the relationship between the mass transfer and properties of single bubbles. The image analysis sequence was applied to calculate the bubble diameters. And the camera was set on a moving platform to be able to record the whole bubble rising process. A mobile bubble surface transits to an immobile surface can be reflected by the transition from high mass transfer rate to the low one. Legendre and Zevenhoven [5] used the similar image analysis sequence to calculate the single bubbles and individual bubbles in small bubble swarms. The bubble behaviors in a tall tower were investigated. Though the theoretical studies about mass transfer were implemented, the experiments or numerical calculations of how the mass transfer influence the rising behavior or vice versa were not enough.

Saito’s research team has focused on the experimental observation of CO2 mass transfer for several years. Saito and Toriu [6] firstly used the LIF/HPTS method to visual the CO2 dissolution. Three sections of rising single bubble were visualized: the linear-ascent, the first-inversion, and the second-inversion of the zigzag motion. The results show that the instantaneous mass-transfer coefficients increased in the linear-ascent section and second-inversion section. Huang and Saito [7] then studied the influence of bubble-surface contamination on instantaneous mass transfer. Compared with the CO2 mass flux in the purified water, that in the contaminated water was two or three times less. On the photos of the bubble, there is a dark region below the bubble and the area of the region in the purified water is larger than that in the contaminated water. Huang and Saito [8] indirectly considered the action of the Marangoni convection and analyzed the forces on the bubbles. Three-dimensional wake structure was obtained from LIF visualization and this makes the mass transfer mechanism clearer [9].

Numerical calculations about CO2 mass transfer can overcome the shortcomings of data acquisition in the experiments. The data can be calculated and obtained directly. In this paper, the CO2 bubble rising with mass transfer in the water was calculated by the computational fluid dynamics software Fluent. VOF method was applied to track the gas-liquid interface. The mass transfer mechanism on the liquid side was implemented by User Defined Function. The bubble dimensions and shapes were calculated and shown. The external flow patterns around the bubble and their influences on the mass transfer process were analyzed.

2. Methodology

2.1. Governing equations

Both the gas and liquid are assumed as incompressible and obeyed the continuity and momentum equations which are shown as Eq.1 and Eq.2:

\[ \nabla \cdot \vec{u} = 0 \tag{1} \]

\[ \frac{\partial (\rho \vec{u})}{\partial t} + \nabla \cdot (\rho \vec{u} \vec{u}) = -\nabla p + \nabla \cdot [\mu (\nabla \vec{u} + (\nabla \vec{u})^T)] + \rho \vec{g} + F_s \tag{2} \]

Where Fs stands for the surface tension source term which is implemented by the continuum surface force (CSF) model. The CSF model correlation is written as Eq.3:

\[ F_s = \sigma \frac{\rho \kappa \nabla \cdot \alpha}{0.5 (\rho_1 + \rho_2)} \tag{3} \]

Where \( \sigma \) is surface tension, \( \kappa = \nabla \cdot \hat{n}, \ \hat{n} = \frac{n}{|n|}, \ n = \nabla \alpha_i \).
VOF method introduced the volume fraction to describe the phases and interface. Take the Fv as the gas phase volume fraction for example, gas phase is determined when the Fv is 1. Liquid phase is determined when the Fv is 0. When Fv is equal to positive values, interphase is determined.

A User Defined Scalar Equation was used in the simulation. The scalar was set as CO$_2$ mass fraction $w$ in the water. The equation was shown as follow:

$$\frac{\partial (\alpha, \rho, w)}{\partial t} + \nabla \cdot (\alpha, \rho, \vec{u}, w - \alpha, D_l \nabla w) = S_i$$

(4)

Where $D_l$ is the diffusion coefficient and $S_i$ is the mass transfer source term. Take the Henry’s law into consideration, the source term can be described as follow [10]:

$$S_i = k_i \alpha (\frac{P}{H} - \rho, w)$$

(5)

Where $H$ is the henry constant, $k_i$ is the mass transfer coefficient.

### 2.2 Mesh geometry and solution methods

The calculation domain in 2D geometry is 50 mm*100 mm, which is large enough to study the CO$_2$ bubble rising with mass transfer. The mesh number is 500000 and it is enough to give satisfactory simulation results. The calculation domain is filled with water initially and the gas is added through an orifice with a diameter of 2.4 mm at the bottom. The gas flux is 10 mL·min$^{-1}$ and the corresponding velocity is 0.037 m·s$^{-1}$. The wall boundary condition is set as no-slip wall and the initial gas velocity is zero.

The pressure-velocity coupling method is PISO scheme and the pressure discretization method is PRESTO! method. The momentum discretization method is QUICK and the volume fraction is discretized by Geo-Reconstruct method. The scalar equation is discretized by First Order Upwind scheme.

### 3. Results and discussion

#### 3.1 Bubble dimension and shapes

The rising process of a CO$_2$ bubble in the water was recorded in Fig.1. The Phase 2 indicated the gas phase. The bubble deforms through the rising process and it rises in a straight-up path initially but then rises along a zigzag path. The rising path is related to the gas and liquid properties, especially for the liquid viscosity. The properties are shown in Table 1.

![Figure 1. The rising process of CO$_2$ bubble.](image)
Table 1. Material properties.

| Material | Molecular Weight (g/mol) | Density (kg/m³) | Viscosity (kg/m·s) | Surface tension (N/m) |
|----------|--------------------------|-----------------|-------------------|---------------------|
| CO₂      | 44                       | 1.7878          | 1.37e-5           | 0.07196             |
| water    | 18                       | 998.2           | 1.003e-3          |                     |

The viscosity of water is only 1.003e-3 kg/m-s so that the bubble shape tends to change when the diameter is large enough. To reflect the bubble deformation, aspect ratio is usually used. It is calculated by the ratio of vertical length and horizontal length of the bubble. The calculation equation is shown as follow:

\[
A_R = \frac{L_y}{L_x}
\]  

where \( A_R \) is the aspect ratio, \( L_y \) and \( L_x \) are the vertical length and horizontal length, respectively.

Figure 2 shows the results of aspect ratio for the single bubble. It should be noticed that the bubble detaches from the orifice after 0.3s so that the aspect ratio can be obtained since then. The aspect ratio fluctuates between 0.27 and 1.07, which agrees with the bubble deformation shown in Figure 1. All the bubble shapes are irregular but the equivalent diameters almost constant at 5.7 mm.

![Figure 2](image-url). The aspect ratio and equivalent diameter of CO₂ bubble.

3.2. Liquid side mass transfer of CO₂ bubble

The bubble behavior can be divided as three sections, which are bubble formation, bubble detachment, and bubble rise.

In the bubble formation section, the bubble was forming at the orifice and the mass transfer process happened. Figure 3a shows the CO₂ mass fraction around the bubble at 0.1s. On the liquid side, the region of CO₂ distributed at the top of the bubble is larger compared with other parts. Figure 3b shows the velocity distribution near the bubble. At the top of the bubble, the velocity is higher, so the large CO₂ region may result from the high velocity. Same phenomenon can be seen in bubble detachment section from Figure 3c and Figure 3d which show the bubble at 0.25s. When the bubble is forming or detaching, the gas from the orifice enters the liquid along the longitudinal and then make the water above the bubble flow quickly. The faster flow velocity makes more fresh water contact with the bubble boundary and then increases the CO₂ mass fraction gradient which intensifies mass transfer.
Therefore, at the top of the bubble, the CO$_2$ mass fraction is higher and the mass transfer of CO$_2$ is better.

In the bubble rise section, the mass transfer conditions are different. Take the bubble at 0.5s for instance, the CO$_2$ transfers mainly from the bottom of the bubble instead of the top (see Figure 3e). There are several CO$_2$ mass fraction wakes occurred below the bubble. Through analyzing the velocity distribution in Figure 3f, it is clear that the wakes are caused by the liquid flow patterns around the bubble.

![Figure 3. CO$_2$ mass fraction and velocity vectors around the bubble: (a)-(b) at 0.1s; (c)-(d) at 0.25s; (e)-(f) at 0.5s](image)

### 4. Conclusion

The rising behavior and the mass transfer process of CO$_2$ bubble in the water were simulated in this paper. Through analyzing the bubble dimension and deformation, the flow patterns around the bubble, and the CO$_2$ mass fraction distribution, the main results are concluded as follow:

1. Though the shapes of CO$_2$ bubble change dramatically, the bubble equivalent diameter keeps constant through the rising process.

2. In the bubble formation and detachment sections, the CO$_2$ transfers better at the top of the bubble because the liquid flows quickly and more fresh water in this region can contact with the bubble to intensify the mass transfer.

3. In the bubble rise section, the main mass transfer region is below the bubble. There are wake structures occurred and the flow patterns influence the CO$_2$ mass fraction distribution.
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