Enhanced Mass Transfer in Microbubble Driven Airlift Bioreactor for Microalgal Culture

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ABSTRACT

In this study, the effect of microfluidic microbubbles on overall gas-liquid mass transfer (CO2 dissolution and O2 removal) was investigated under five different flow rates. The effect of different liquid substrate on CO2 mass transfer properties was also tested. The results showed that the $K_La$ can be enhanced by either increasing the dosing flowrate or reducing the bubble size; however, increasing the flow rate to achieve a higher $K_La$ would ultimately lower the CO2 capture efficiency. In order to achieve both higher CO2 mass transfer rate and capture efficiency, reducing bubble size (e.g. using microbubbles) has been proved more promising than increasing flow rate. Microbubble dosing with 5% CO2 gas showed improved $K_La$ by 30% - 100% across different flow rates, compared to fine-bubble dosing. In the real algal culture medium, there appears to be two distinct stages in terms of $K_La$, divided by the pH of 8.4.

Keywords: Microbubbles; $K_La$; CO2 Capture; Algal Culture

1. Introduction

The cultivation of microalgae has been studied and developed for more than 40 years [1]. Two of the major limiting factors for microalgal culture are light and CO2 as they are the key participants for the “light reactions” and “dark reactions” in photosynthesis, respectively. Many researches have been carried out to study the impact of light on algal growth. Technical issues associated with light have been also well studied especially for photobioreactors, with various solutions (e.g. using an optimal mixing rate and light/dark ratio, combining artificial light with natural light, and increasing harvest frequency etc.) [2]. As regards to CO2 supply, in most microalgal cultures, CO2 is usually injected into the culture through bubbling CO2 enriched air into porous diffusers, which promises a gas transfer efficiency of 13% - 20% [3]. Additional supply of CO2 contributes many benefits to the culture. First of all, the supply of CO2 can lead to enhanced algal metabolisms, and on the other hand, it can act as buffer solution to neutralize the increased pH caused by algal growth. Secondly, supply of CO2 enhances the internal mixing of bioreactor, helping to evenly distribute nutrients and the exposure time of algal cells to light. Furthermore, accumulation of O2 in culture medium is toxic to microalgal cells and it is one of the major limiting factors for scale up of the bioreactor [4]. Introducing CO2 into culture also helps to strip accumulated oxygen and hence prevents algal cells from toxicity [5]. According to the relationship between partial pressure and Gibbs free energy (Equation (1)), it is found that the increase in the partial pressure of reactants (e.g. CO2) or the reduction of partial pressure in the products (e.g. O2) results in the value of Gibbs free energy becoming negative. Hence the reaction becomes thermodynamically favourable and moves towards to the formation of more products [6]. Such feature of performance is widely utilized for many bioprocesses to achieve a higher productivity [2,5]. Therefore by increasing the concentration of dissolved CO2 whilst reducing the accumulated O2 level can be considered as an approach towards improving productivity.

\[ aA + bB \rightarrow cC + dD \]

\[ \Delta G = \Delta G^r + RT \ln \frac{[p_C]^c [p_D]^d}{[p_A]^a [p_B]^b} \]  

(1)
However, most existing CO₂ supply techniques are relatively inefficient. Due to low interfacial surface area between gas bubbles and culture medium, the gas-liquid mass transfer is poor, which associated with CO₂ loss to atmosphere [7]. Besides, additional CO₂ supply increases the operational cost, which cannot be balanced eventually by the algal yields enhancement due to the low CO₂ mass transfer. Improving the CO₂ supply efficiency and consequently enhancing the algal productivity has become a major challenge over the years. Design of bioreactor with low energy cost and particularly high gas mass transfer for both CO₂ dissolution and O₂ removal is hence a major consideration for cost-competitive microalgae culture.

Due to the enhanced gas-liquid mass transfer efficiency and liquid circulation etc., airlift bioreactors (ALB) are increasingly employed for microalgae culture. Many researches have been carried out on the performance of different ALBs; however, these studies were carried out all based on conventional gas supply system. There are few studies on the effects of microbubbles on ALB performance, because normally the microbubble generation systems, for instance DAF, electro-flotation, electrostatic spraying, and mechanical agitation etc, were not profitable to be applied into most bio-processes due to their high energy cost [8–12]. Recently, an innovative microbubble generation system (fluidic oscillator) with lower power consumption has been invented by Zimmerman et al. [13,14] with the benefits of energy saving and improved efficiency. The detailed information on fluidic oscillator and its microbubble generation mechanism were described in previous studies [13-15]. This study aims to investigate the effect of microbubbles (generated by fluidic oscillator) on mass transfer under different aeration flow rates. In addition, the impact of different liquid substrate (e.g. NaHCO₃ medium and algae medium) on CO₂ mass transfer properties is investigated.

2. Materials and Methods

2.1. Materials

A 7L-aerifft loop bioreactor based on classic ALB geometry designs [16], as shown in Figure 1 (left), was used to study the mass transfer properties of microbubbles and fine-bubbles. Besides, a smaller version (3L) of ALB, based on the similar geometry design, was applied to study the impact of different liquids on mass transfer, shown in Figure 1 (right).

2.2. Experimental Procedure

To study the mass transfer properties of microbubbles and fine-bubbles, the two inlet ports of diffusers at the bottom of bioreactor were connected to gas cylinder by PVC tubes, through a fluidic oscillator or a Y-junction.

The 3L-ALB airlift bioreactor is illustrated in Figure 1 (right). A pH and DO probe (Mettler Toledo, UK) were inserted into the bioreactor via the holes on the lid. These holes were blocked by rubber bungs to prevent gas leakage. The outlet nozzle on the lid was connected to a flow meter to measure the outlet flowrate which is equal to the real inlet flowrate. For each set of experiment, 7L distilled water with the temperature 25°C ± 1°C were employed. Mixture gas containing 5% CO₂ and 95% N₂ was injected into bioreactor under certain flow rate. Five different flow rates were tested. The flow rate was measured by a flow meter which was connected to the outlet port of the bioreactor. The changes in pH and DO were monitored by pH meter and DO meter respectively. Data was recorded every 30 seconds until pH and DO readings were stable. The effect of different liquids on mass transfer was studied in the 3L-ALB with the same setup as shown in Figure 2. The mass transfer for CO₂ dissolution was tested in the distilled water containing certain concentration of NaHCO₃ and also in the real algal culture medium (containing algae). 7 different concentrations of NaHCO₃ were tested. The algae (Dinofalicia salina) used in this study was 7 days old. During the mass transfer test, 5% CO₂ and 95% N₂ was injected into D. Salina culture under a fixed dosing flow rate (0.7L/min), with DO and pH recorded every 30 seconds. The dissolved CO₂ concentration was calculated based on Equation (2) (for water).
or Equation (3) (for NaHCO₃ medium and algal medium) [17]. [Na⁺] in Equation (3) particularly means the concentration of Na⁺ obtained from NaHCO₃. The method of mass transfer coefficient estimation was estimated as the slope of a semilog plot of 1/(1-E) versus T, which was mass transfer coefficient estimation was estimated as the total interfacial area was amplified.

3. Results and Discussions
3.1. Mass Transfer for Microbubble Driven and Fine-Bubble Driven Reactor

The effects of microbubble dosing on mass transfer for CO₂ dissolution and O₂ removal were examined by dosing 5% CO₂ mix-gas (balanced with 95% N₂) into bioreactor (containing 7L distilled water) under 5 different bubbling flow rates, along with the control experiment (without fluidic oscillator, fine bubble dosing). The mass transfer coefficient \( K_{La} \) for CO₂ dissolution and O₂ removal under each bubbling condition were plotted in Figure 3. From Figure 3, generally \( K_{La} \) for either CO₂ dissolution or O₂ stripping increases along with gas dosing flow rate. For \( K_{La} \), \( K_i \) mainly depends on the gas-liquid properties (e.g. density, viscosity, diffusivity and temperature etc.), and therefore is usually considered as a constant for the fixed circumstance [16]. Chisti expressed the interfacial area “a” as a function of gas holdup (e) and bubble diameter \( d_B \) [16], shown as:

\[
a = \frac{6e}{d_B}
\]

(4)
reducing the bubble size can lead to an improvement on $K_{La}$ as well. In another word, $K_{La}$ can be enhanced by either increasing the dosing flowrate (to be more accurate, flowrate/liquid volume ratio) or reducing the bubble size.

3.2. The Improvement of $K_{La}$ by Using Fluidic Oscillator

When injecting CO$_2$/N$_2$ mixture gas into water, CO$_2$ dissolution happens along with O$_2$ stripping. The improvements by using fluidic oscillator (microbubbles) on mass transfer for CO$_2$ dissolution and O$_2$ stripping can be simply quantified as the percentage increase in $K_{L_a}$ for CO$_2$ and $K_{L_a}$ for O$_2$, expressed in Equation (5) and Equation (6), respectively.

\[
I_{K_{L_a}}^{\text{CO}_2}\% = \frac{K_{L_a}^{\text{CO}_2} - K_{L_a}^{\text{NOFO}}}{K_{L_a}^{\text{NOFO}}} \tag{5}
\]

\[
I_{K_{L_a}}^{\text{O}_2}\% = \frac{K_{L_a}^{\text{O}_2} - K_{L_a}^{\text{NOFO}}}{K_{L_a}^{\text{NOFO}}} \tag{6}
\]

Either Equation (5) or Equation (6) can be turned into Equation (7) which indicates the percentage increase in $K_{La}$ by using microbubble dosing should be the same for either CO$_2$ dissolution or O$_2$ removal under a fixed bubbling flowrate. The percentage improvement of $K_{La}$ is therefore determined by the percentage difference of the total interfacial areas for a certain dosing flow rate. Combining Equation (7) and Equation (4), the percentage increase in $K_{La}$ is correlated to bubble diameter ($d_b$) and gas hold-up ($\varepsilon$), described by Equation (8).

\[
I_\% = I_{K_{L_a}}^{\text{CO}_2}\% = I_{K_{L_a}}^{\text{O}_2}\% = \frac{\varepsilon_{\text{FO}} d_b - \varepsilon_{\text{NOFO}} d_b}{\varepsilon_{\text{NOFO}} d_b} \tag{7}
\]

\[
I_\% = \frac{\varepsilon_{\text{FO}} - \varepsilon_{\text{NOFO}}}{\varepsilon_{\text{NOFO}}} 
\]

From Equation (8), the efficiency of $K_{La}$ improvement (1%) therefore should be the same across different flow rates, assuming 1) the gas holds are identical between microbubble dosing and fine bubble under the same flow rates, and 2) changing the flow rate dose not vary the average bubble size for either microbubbles or fine bubbles as long as the “bubble coalescence” does not happen. However, the experimental results are inconsistent with such speculation. Figure 4 shows the $K_{La}$ percentage increase. In general, microbubble dosing enhances the $K_{La}$ by 30% - 100% over a wide flow rate range, while the efficiency of the improvement decreases when increasing the flow rate. It is speculated that the microbubble size increases with the flow rate. The fluidic oscillator provides a periodical oscillating pulse to neck-off the bubbles attached to the diffuser orifice when they are still small. But for the same surface area of diffuser, increasing the flow rate may change the oscillating properties (e.g. the attenuation of “pulse force” due to the build up of boundary layer), and may also cause bubble coalescence, consequently weakening the efficiency of oscillator for microbubble creation. Therefore, the microbubble size may slightly increase when the flow rate increases, resulting in a reduction of $d_{\text{NOFO}}/d_{\text{FO}}$ ratio, which leads to the decline of $K_{La}$ improvement efficiency ($I_\%$). This phenomenon also indicates a view that using fluidic oscillator to enhance mass transfer has its limitations in terms of flow rate (or to be more accurate, flow rate over liquid volume ratio).

3.3. The Relationship between Mass Transfer Coefficient and Overall Mass Transfer Rate

Knowing the mass transfer coefficient $K_{La}$ helps to indicate the capability of mass transfer, while knowing the mass transfer rate gives a straight view of e.g. how rapidly the CO$_2$ dissolve into liquid, which also helps to estimate the CO$_2$ capture efficiency.

The instantaneous mass transfer rate ($V_{MTR}$) is interpreted as the driving force multiplied by the mass transfer coefficient ($K_{La}$) [16], shown in Equation (9),

\[
V_{MTR} = \frac{d[\text{CO}_2]}{dt} = K_{La}[[\text{CO}_2]^* - [\text{CO}_2]] \tag{9}
\]

where $K_{La}$ is the mass transfer coefficient (min$^{-1}$), both $[\text{CO}_2]$ and $[\text{CO}_2]^*$ are instantaneous concentrations of CO$_2$ and its equilibrium concentration (mol·L$^{-1}$), respectively. The average mass transfer rate ($V'_{MTR}$) for a certain dosing time period ($t_d$) can be fairly represented as

\[
V'_{MTR} = \frac{\int_{t_0}^{t_d} V_{MTR} dt}{t_d} = \int_{t_0}^{t_d} \frac{K_{La}([[\text{CO}_2]^* - [\text{CO}_2]]) dt}{t_d} \tag{10}
\]

Assuming

\[
[\text{CO}_2] = [\text{CO}_2]_0 + V'_{MTR} t \tag{11}
\]

by solving Equation (10) and Equation (11), it gives:
where $[CO_2]_0$ represents the initial CO2 concentration (mol·L$^{-1}$) for a selected time period.

The accuracy of Equation (12) was examined via Figure 5 which plots the experimental values of average mass transfer rates versus the calculated values by using Equation (12). Compared with examined values, most of the data calculated by Equation (12) showed only less than 10% difference.

3.4. CO2 Capture Efficiency for Microbubble Dosing and Fine-Bubble Dosing

CO2 capture efficiency is one of the most important parameters concerned by many bioprocesses with the purpose of CO2 sequestration. Since the rate of CO2 dissolving into liquid can be evaluated by overall mass transfer rate using Equation (12), the CO2 capture efficiency $E_{CO_2}$ can be therefore simply described as the amount of CO2 been absorbed over the amount of CO2 been fed into the liquid ($m_{CO_2}/m_{CO_2}$) within a specific dosing time period ($t_d$), shown in Equation (13).

$$E_{CO_2} = \frac{m_{CO_2}}{m_{CO_2}} = \frac{V'_{MTR} \times Vol \times t_d}{CO_2 \% \times \eta \times U \times (RT) \times t_d}$$  \hspace{1cm} \text{(13)}$$

where CO2% means the percentage of CO2 in the gas supply, Vol is the volume of the liquid (m$^3$), $V_F$ is the gas dosing flow rate (L·min$^{-1}$), P is standard atmosphere pressure (101,325 Pa), R is the ideal gas law constant (8.314 J·K$^{-1}$·mol$^{-1}$) and T is the temperature (298 K).

The CO2 dissolving rate and the CO2 capture efficiency under different dosing conditions were plotted in Figures 6 and 7, respectively. In general, micro- bubble dosing by using the fluidic oscillator was found to have both higher CO2 dissolving rate (average mass transfer rate) and CO2 sequestration efficiency for a wide range of dosing flow rate, but the level of improvements were attenuated as the flow rate went up (similar to the attenuation of $K_{La}$ improvement, see 3.2). Such attenuation of improvement was caused by the increase in microbubble size due to the weakening of oscillation and bubble coalescence under higher flow rate.

Apart from reducing bubble size, increasing flow rate can also achieve a higher $K_{La}$ (see 3.1), it is therefore not a surprise to found that the CO2 overall mass transfer rate increases along with the flow rate (Figure 6). However, it is interesting that the CO2 capture efficiency actually reduces when the flow rate increases (Figure 7). Higher $K_{La}$ dose mean higher CO2 overall mass transfer rate (higher CO2 dissolving rate), however, if the cost to achieve higher $K_{La}$ is enhancing the dosing flow rate rather than reducing bubble size, then the amount of not dissolved CO2 (“wasted CO2”) would increase, and such increase in wasted CO2 could not be balanced by the increase in dissolved CO2, which ultimately lowers the CO2 capture efficiency. Therefore, in order to achieve both higher CO2 mass transfer rate and capture efficiency, reducing bubble size (e.g. using microbubbles) is more promising than increasing flow rate.
3.5. Effect of NaHCO₃ on Equilibrium pH and CO₂ Mass Transfer Rate in Water

In microalgae culture, CO₂ is injected into the culture medium (usually containing NaHCO₃) rather than pure water. When adding NaHCO₃ into water, NaHCO₃ dissociates into sodium (Na⁺) and bicarbonate (HCO₃⁻) ions, and these HCO₃⁻ ions neutralize some of the H⁺ ions present in the medium to form the dissolved CO₂ and so increase the pH. So the concentration of NaHCO₃ clearly has an effect on pH, it is worth finding out whether the culture medium containing NaHCO₃ could affect the CO₂ mass transfer. Therefore, a separate experiment was carried out in a smaller version but the same design of airlift bioreactor (2.5 L).

Keeping other parameter constant (flow rate, temperature etc.), higher concentrations of NaHCO₃ added into distilled water should theoretically minimal pH (equilibrium pH, pH*) reached after CO₂ dosing. According to Henry’s law and Two-film theory, the equilibrium concentration of dissolved CO₂ ([CO₂]*) should only depend on the CO₂ partial pressure in the gas phase for a fixed gas/liquid properties and temperature (assuming the changes in liquid physical properties by adding different amount of NaHCO₃ to the water, e.g. viscosity, are negligible, as long as the concentration of NaHCO₃ is low). Therefore, different concentrations of NaHCO₃ in the water should not affect the [CO₂]*. On the other hand, the CO₂ concentration is correlated to pH by Equation (3). Since the concentration of Na⁺ varies for different concentration of NaHCO₃, while the [CO₂]* does not change, it is therefore reasonable that pH* changes for the water containing various NaHCO₃ concentration. Indeed, this hypothesis was proved correct, in Figure 8, a log-linear trend was observed in the equilibrium pH values as the concentration of NaHCO₃ was increased. Besides, all the equilibrium concentrations of CO₂ corresponding to each equilibrium pH value under different concentrations of NaHCO₃ were found to be the same, which is approximately 0.0017 ± 0.0001 mol·L⁻¹. In terms of mass transfer for CO₂ dissolution, it can be seen that changing the concentration of NaHCO₃ does not have much of an effect on the mass transfer coefficient (Figure 9). Hence, NaHCO₃ could be used to control the equilibrium (minimal) pH of the medium without affecting the [CO₂]* and KLa. The pH region can also be altered depending on the particular strain of microalgae being cultured, as different algae prefer different pH.

3.6. CO₂ Mass Transfer in Microalgae Culture

In order to test the effect of real microalgae culture on CO₂ mass transfer, 5% CO₂ was dosed into a healthy D. Salina culture (containing 0.012 mol/L of NaHCO₃) under a fixed flow rate (0.7 L/min) for 30 min, with pH recorded every 30 seconds. The results showed that there appears to be two distinct stages in terms of KLa, see Figure 10 for example. The calculations leading to the determination of the KLa mass transfer coefficients from the slopes seen in Figure 10 are given in Table A1 (See Appendix).

From the calculations in Table A1, for the first 4.5 minutes of gas supply, the concentration of CO₂ dissolved and the resultant mass transfer is of different magnitudes when the pH is greater than 8.4. Once the pH is less than 8.4, the KLa is much higher in comparison. This was observed for each mass transfer test in culture medium with the threshold pH value of 8.4 seen each time. It is speculated that slower mass transfer at the start when pH is higher than 8.4 could be due to the chemical reactions taking place within the culture medium the gas is being bubbled through. Considering the dissociation of water into hydrogen (H⁺) and hydroxyl (OH⁻) ions, when the pH is over 8.4, the concentration of hydroxyl ions will be much greater than that of the hydrogen ions ([OH⁻] << [H⁺]). The [H⁺] produced when CO₂ dis-
pose, at pH > 8.4 the K_{i,a} for dissolved CO_{2} is relatively low, but it dose not mean less CO_{2} from gas supply has been transferred into liquid. Actually, most of the CO_{2} been transferred into culture was converted into HCO_{3}^{-} and CO_{3}^{2-} when pH > 8.4. Therefore, when calculating the CO_{2} capture efficiency in future, the changes in the amount of total carbon (C_{T}) should be considered rather than the amount of dissolved CO_{2} when pH > 8.4. But if the pH is less than 8.4, the changes in the amount of total carbon almost come from the changes in dissolved CO_{2}, so it is fair to use the K_{i,a} of dissolved CO_{2} for CO_{2} capture efficiency estimation.

Comparing both CO_{2} mass transfer (when pH < 8.4) under 0.7 L·min^{-1} of dosing for water containing NaHCO_{3} and the culture medium including microalgae (with the same concentration of NaHCO_{3}), the K_{i,a} in water (0.2531 min^{-1}) was found to be greater than the one in the presence of D. salina (0.1776 min^{-1}). That may be because the cells in the medium increased its viscosity, which could have reduced the diffusivity of CO_{2} from liquid film to liquid phase plus part of dissolved CO_{2} could be consumed due to D. salina growth. Hence the rate of CO_{2} diffusion into the culture was slowed down, whilst without D. salina present the CO_{2} could diffuse much easier through the medium and without being consumed. Also, because of the changes in liquid properties, the CO_{2} equilibrium concentration [CO_{2}]^{*} was slightly smaller in the culture (0.0011 ± 0.0001 mol·L^{-1}) than that in the NaHCO_{3} medium (0.0017 ± 0.0001 mol·L^{-1}).

4. Conclusions

For the same bubble generation method, enhancing the gas dosing flow rate can increase the mass transfer coefficient. For the same bubbling flow rate, reducing the bubble size can lead to an improvement on K_{i,a} as well. Compared with fine-bubble dosing, microbubbles dosing of 5% CO_{2} gas by using fluidic oscillator has been proved to enhance the K_{i,a} for both CO_{2} dissolution and O_{2} removal by 30% - 100% across different flow rate. Despite K_{i,a} can be enhanced by either increasing the dosing flowrate (to be more accurate, flowrate/liquid volume ratio) or reducing the bubble size, increasing flow rate to achieve a higher K_{i,a} would also raise the amount of CO_{2} being wasted (not dissolved) which would ultimately lower the CO_{2} capture efficiency. Therefore, in order to achieve both higher CO_{2} mass transfer rate and capture efficiency for the improvement of microalgal growth and CO_{2} sequestration, reducing bubble size (e.g. using microbubbles) is more promising than increasing flow rate.

The K_{i,a} for CO_{2} dissolution was not affected by the presence of NaHCO_{3}, and NaHCO_{3} could be used to control the equilibrium pH of the medium without af-

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**Figure 9. Changes in CO_{2} mass transfer coefficients for different concentrations of NaHCO_{3}**

The slope of straight line indicates mass transfer coefficient K_{i,a} (min^{-1}).

**Figure 10. Typical plot for K_{i,a} estimation (for 0.7 L·min^{-1} dosing).**
fecting the $[\text{CO}_2]^*$ and $K_{La}$. The pH region can also be altered depending on the particular strain of microalgae being cultured, as different algae prefer different pH. In the real algal culture, there appears to be two distinct stages in terms of $K_{La}$ divided by the pH of 8.4. When pH < 8.4, due to the changes in liquid properties and carbon system equilibrium relations, the $K_{La}$ as well as $[\text{CO}_2]^*$ was found slightly reduced than the ones in the water.

Future works need to be done to test the effect of different percentage of CO$_2$ in the gas supply on mass transfer. And a mathematical model correlating mass transfer to bubble size, flow rate/liquid volume ratio and percentage of CO$_2$ in the gas supply etc. is expected to be established, which could facilitate the estimation of CO$_2$ dosing time for microalgal culture.

5. Acknowledgements

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REFERENCES


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Table A1. An example of calculations leading to the CO₂ mass transfer coefficient (for 0.7 L·min⁻¹ dosing)

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<th>Time (s)</th>
<th>pH</th>
<th>[CO₂] (Equation (3)) (mg·L⁻¹)</th>
<th>ln([CO₂]² − [CO₂]₀)/([CO₂]² − [CO₂]ₜ)</th>
<th>KLa  (min⁻¹)</th>
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