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Efficiency of CO_2 fixation by microalgae in a closed raceway pond

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HIGHLIGHTS

- A closed raceway pond and its equation of CO_2 mass transfer.
- CO_2 dissolution efficiency in the closed raceway pond without algae.
- CO_2 fixation efficiency by algae with continuous aeration in the pond.
- A mode of intermittent aeration with CO_2 fixation efficiency of 95% in the pond.

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ABSTRACT

Microalgae contain about 50% of carbon, which means that a total of 1.83 ton of CO_2 is needed to produce 1 ton of microalgae. The cost of CO_2 supply for microalgal large scale cultivation should be considered and the low CO_2 fixation efficiency by microalgae will lead to much more expenditure of CO_2. In this study, a closed raceway pond was constructed by covering a normal open raceway pond with a specially designed transparent cover, which directly touched the surface of microalgal culture media. This cover prevented supplied CO_2 escaping into atmosphere and thus increased the retention time of CO_2. The CO_2 gas–liquid mass transfer and CO_2 fixation efficiency by microalgae in the closed raceway pond were investigated, and the model of CO_2 fixation by microalgae was developed. Through the model, the CO_2 fixation efficiency increased to 95% under intermittent gas sparging.

1. Introduction

Microalgae have received extensive research for five decades because of their possible commercial application in biofuels, cosmetics, pharmaceuticals, nutrition and food additives, aquaculture, pollution prevention, wastewater treatment and so on (Mata et al., 2010; Milledge, 2010; Spolaore et al., 2006). However, the high cost of microalgal cultivation is one of the greatest obstacles for commercial application. The availability and cost of CO_2 are major factors in the economics of algal mass culture. It was estimated that the cost of carbon source ranged from about 8% to 27% of the production costs of the algae (Laws and Berning, 1991). The cost of carbon source will be multiplied with the lower CO_2 fixation efficiency. There are many factors influencing CO_2 fixation efficiency in microalgal cultivation, including microalgal strains, structures of photobioreactors, light quantity and quality, operating models and nutrition conditions. The structures of photobioreactors have an important effect on CO_2 fixation efficiency because they affect CO_2 dissolution efficiency and CO_2 utilization efficiency by microalgae. Open-culture systems normally cannot use supplied CO_2 effectively, which easily escaped from the culture media due to the shallow depths and poor CO_2 mass transfer efficiency, and CO_2 fixation efficiency ranged from 10% to 30% (Becker, 1994; Weissman and Goebel, 1985). This low efficiency results in microalgal cultivation cost intensive (Campbell et al., 2011). In order to improve CO_2 fixation efficiency by microalgae in open raceway pond, Ketheesan and Nirmalakhandan (2012) designed an airlift-driven raceway reactor for microalgal cultivation with the maximum CO_2 utilization efficiency of 33%, and Putt et al. (2011) designed and investigated CO_2 transfer efficiency of a carbonation column combining with open ponds without analyzing the energy consumption and the microalgal productivity in the photobioreactor. Compared with open culture systems, closed photobioreactors could reduce CO_2 losses because they can prolong CO_2 retention time and improve mass transfer efficiency. Many researchers have studied microalgal CO_2 fixation with closed photobioreactors (Chiu et al., 2008; de Morais and Costa, 2007; Cheng et al., 2006; Keffer and Kleinheinz, 2002). But the CO_2 fixation efficiency was not high,

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especially for the high concentration of CO_2. Chiu et al. (2008) reported that the efficiency of CO_2 removal by *Chlorella* sp. was 16% at CO_2 concentration of 15%, and in treatment of 2% CO_2 gas sparging, CO_2 fixation efficiency was 58% in a semi-continuous photobioreactor. de Morais and Costa (2007) reported that the CO_2 fixation efficiency was lower with high CO_2 concentration (12%) than that with low concentration (6%). In the present study, a closed raceway pond was proposed and its structure can increase the CO_2 retention time. Cheng et al. (2006) had demonstrated that increasing retention time of CO_2 in photobioreactor will significantly enhance the CO_2 fixation efficiency. The objective of this work was to investigate the CO_2 fixation efficiency by microalgae at high CO_2 concentration in the closed raceway pond and develop dynamic model of CO_2 fixation, which is used to provide method for the improvement of CO_2 fixation efficiency.

2. Methods

2.1. Closed raceway pond (CRWP)

The schematic diagram of the closed raceway pond is shown in Fig. 1. The CRWP was a four-channel raceway-type pond, being 1.2 m long, 0.25 m wide and 0.09 m deep, with each channel 0.06 m width. The CRWP had two parts and was made of polymethyl methacrylate. The bottom part of the CRWP was a rectangular box without top surface, and the cover part was H-shaped with the water clapboards attached to the lower surface of the cover. A paddle wheel was used for mixing and stirring of the culture medium. The enriched CO_2 gas was aerated into the CRWP by a porous tube from CO_2 inlet. As Fig. 1 shows, the H-shaped cover directly touched the culture liquid surface and kept gas bubbles going along with culture flow. The dashed line in Fig. 1 represents the surface of culture liquid, which touched the cover. The culture liquid was 0.04 m deep. The outlet of exhausted gas in CRWP was in the paddle wheel mixing zone. The arrows in channels represent the direction of culture flow.

2.2. Microalgal cultivation

Strain *Chlorella vulgaris* was obtained from College of Marine Life Sciences, Ocean University of China. Algal cells were cultivated in the following medium (per liter) (Li et al., 2012), containing 1000 mg KNO_3, 237 mg KH_2PO_4, 88 mg $CaCl_2{\cdot}2H_2O$, 40 mg EDTA, 30 mg $FeSO_4{\cdot}7H_2O$, 204 mg $MgSO_4{\cdot}7H_2O$, and 1 mL of trace metal solution. The trace metal solution (per liter) included 0.83 g H_3BO_3, 0.17 g $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$, 0.51 g $CoCl_2{\cdot}6H_2O$, 3.3 g $MnCl_2{\cdot}4H_2O$, 0.95 g $CuSO_4{\cdot}5H_2O$, 2.7 g $ZnSO_4{\cdot}7H_2O$. The flow velocity of culture medium in CRWP was 0.03 m s^{-1} with 12 L working volume. Culture was aerated continuously (or intermittently) with 15% CO_2 at different gas flow rates, and the gas flow rate was measured using a quality flow meter (LZB-3, Yinhuan, China). The microalgal cells were incubated at 24 ± 1 °C under the light intensity of 100 μmol m^{-2} s^{-1} with continuous light, and the light intensity was determined using a light meter (Hansatech Instruments Quantitherm light meter thermometer, Norfolk, England). The pH value of culture was measured using a pH meter (PB-10, Sartorius, Germany) and the concentration of dissolved oxygen (DO) in culture was determined using a DO meter (M700, Mettler Toledo, Switzerland). The dissolved CO_2 concentration in media was determined by a CO_2 meter (FC-200, Shanghai su park information technology Co. Ltd., China).

Culture suspension with a volume of 30 mL was filtered through pre-dried and pre-weighted 2 μm membrane filters, after filtration the filter was rinsed with distilled water, and the filter with microalgal cells was dried at 105 °C in an oven overnight. The filter was removed from the oven, kept in a desiccator and reweighted. This weight minus the weight of the empty filter gave the biomass density. Samples were generally run in triplicate for each data point. The biomass productivity (P, g L^{-1} d^{-1}) of microalgae was calculated from the variation in biomass density (X, g L^{-1}) according to the equation $P = \frac{X_t - X_0}{t - t_0}$, where X_0 was the initial biomass density at time t_0 and X_t was the biomass density at any time t subsequent to t_0.

2.3. Calculation of CO_2 volumetric mass transfer coefficient

The CRWP containing 12 L media without microalgal cultivation was aerated with different concentrations of CO_2 (1.2%, 2%, 15%, 100% (v/v) with N_2 balance) for 120 min at 24 ± 0.5 °C. The liquid velocity (v_{wat}) was 0.03 m s^{-1} and the dissolved CO_2 concentration in culture media was recorded every five minutes. According to two-film theory (Carvalho et al., 2006) and Henry's law, the volumetric mass transfer coefficient of CO_2 in media was calculated from the following equation:

$$\frac{dC}{dt} = K_L a(C^* - C) \tag{1}$$

$\frac{dC}{dt}$, volumetric transport rate of CO_2 in liquid (mol min^{-1} L^{-1}); K_La, volumetric mass transfer coefficient of CO_2 (min^{-1}); C^*, concentration of CO_2 in culture in equilibrium with CO_2 content in CO_2 bubbles (mol L^{-1}), which can be determined via Henry's Law; C, CO_2 concentration in culture media (mol L^{-1}).

Integration for $C = C_0$ at $t = 0$, led to the following equation:

$$\ln\frac{C^* - C}{C^* - C_0} = -K_L a t$$

A plot of left hand side of this equation against time was used to obtain $-K_La$ as the slope.

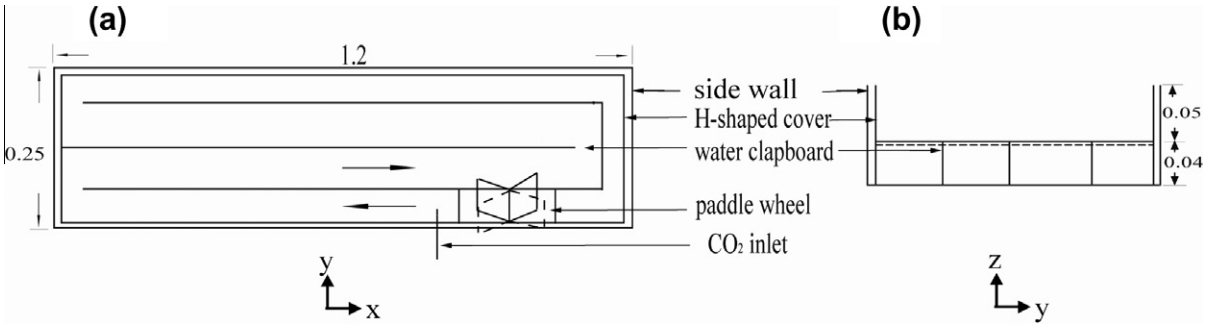

Fig. 1. The schematic diagram of the closed raceway pond (a): top view of the closed raceway pond; (b): side view of the closed raceway pond.

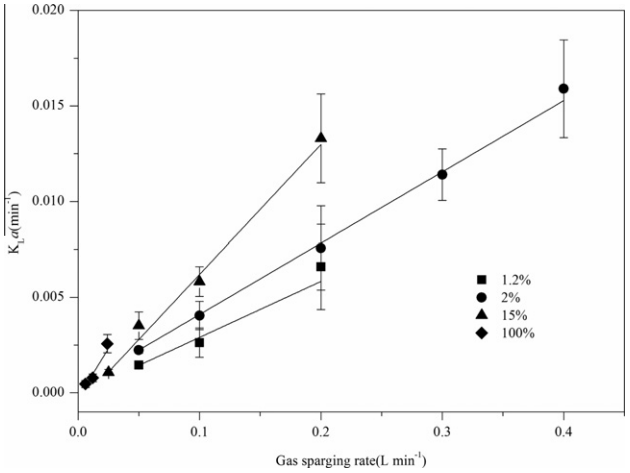

Fig. 2. The relation between gas sparging rate and K_La.

3. Results and discussion

3.1. CO_2 volumetric mass transfer and dissolution efficiency in CRWP

The CO_2 volumetric mass transfer coefficients of different concentrations of CO_2 in culture media in CRWP aerated with different rates are shown in Fig. 2. With the same gas sparging rate, K_La increased with the increase of CO_2 concentrations. Under the same CO_2 concentration, K_La was proportional to gas sparging rate. A linear regression equation between K_La and gas sparging rate (Q) can be obtained:

$$K_La = AQ \tag{2}$$

where Q is the volumetric flow rate of gas ($L\,min^{-1}$) and A is a coefficient.

The A value at CO_2 concentrations of 1.2, 2, 15, and 100% was 0.031 (R^2 = 0.9894), 0.039 (R^2 = 0.9991), 0.065(R^2 = 0.9944), 0.097 (R^2 = 0.9568), respectively. Integration of Eqs. (1) and (2) led to:

$$\frac{dC}{dt} = A(C^* - C)Q \tag{3}$$

The transport rate of CO_2 in CRWP aerated with different gas flow rates and CO_2 concentrations can be calculated from Eq. (3).

According to Eq. (3), when the change of $(C^* - C)$ can be neglected in short time, the number of mole of dissolved CO_2 (n_{CO_2-dis}, mol) in culture media can be expressed as follows:

$$n_{CO_2-dis} = A(C^* - C)QtV$$

where t is the gas sparging time (min) and V is the volume of culture media (L).

The number of mole of CO_2 supply (n_{CO_2-sup}, mol) can be determined from the following equation:

$$n_{CO_2-sup} = \frac{Qtp}{M},$$

where p is the CO_2 concentration, and M is the molar volume of CO_2 ($mol\,L^{-1}$) and is calculated by the following equation:

$$M = \frac{22.4 \times (273 + T)}{273}$$

where T is the culture temperature (°C).

The calculated CO_2 dissolution efficiency (η) in CRWP was obtained:

$$\eta = \frac{n_{CO_2-dis}}{n_{CO_2-sup}} = \frac{A(C^* - C)VM}{p} \tag{4}$$

According to Eq. (4), the CO_2 dissolution efficiency in CRWP without microalgae can be calculated. Fig. 3 shows the relationship of dissolved CO_2 concentration in media and CO_2 dissolution efficiency. The maximal mean relative error between the experimental

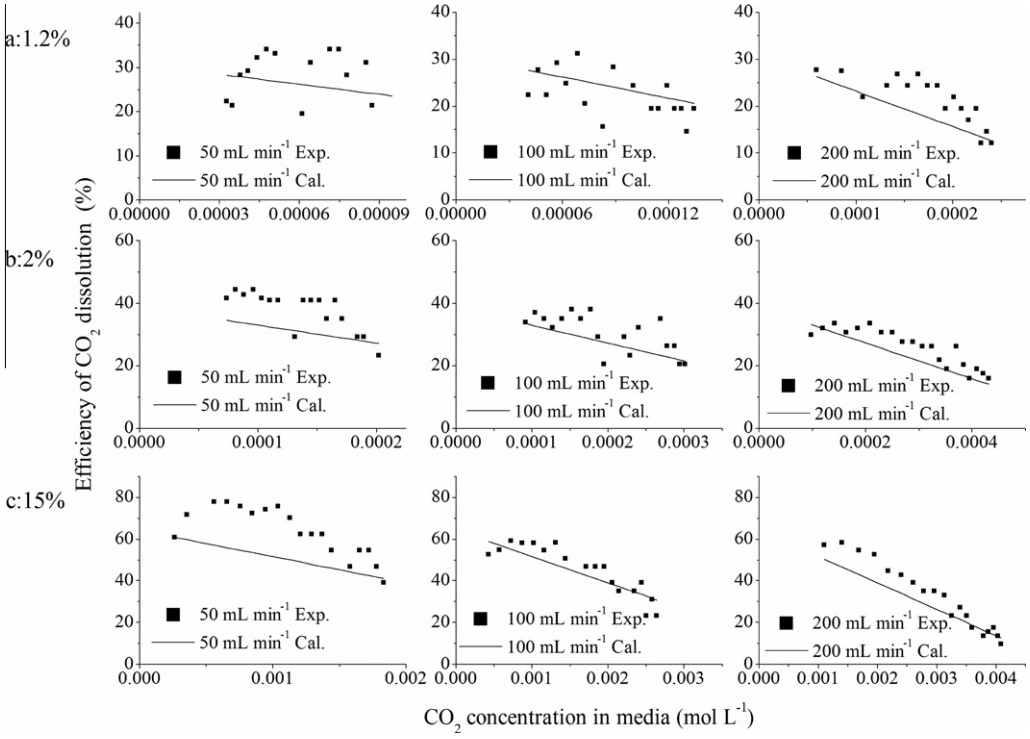

Fig. 3. Experimental and calculated values of CO_2 dissolution efficiency in CRWP. Exp. and Cal. represent experimental and calculated values, respectively. The experimental values were determined from the following equation: $\frac{C_{var}V}{n_{CO_2-sup}}$, where C_{var} was the variation of CO_2 concentration in media in five minutes ($mol\,L^{-1}$).

Table 1
Microalgal productivity and CO_2 fixation efficiency with different gas flow rates.

Gas flow rate ($L\ min^{-1}$)	Microalgal productivity ($g\ d^{-1}\ L^{-1}$)	CO_2 fixation efficiency (%)[a]
0.025	0.2349 ± 0.0168	53
0.035	0.3496 ± 0.0117	56
0.05	0.3219 ± 0.0109	36

[a] The CO_2 fixation efficiency was calculated as follows: $\frac{0.5PV}{12\times60\times24\times n_{CO_2-sup}}\times 100\%$, where 0.5, 12, 60 and 24 represent carbon percentage content in dried microalgae, carbon molecular weight, minutes in an hour and hours in a day, respectively.

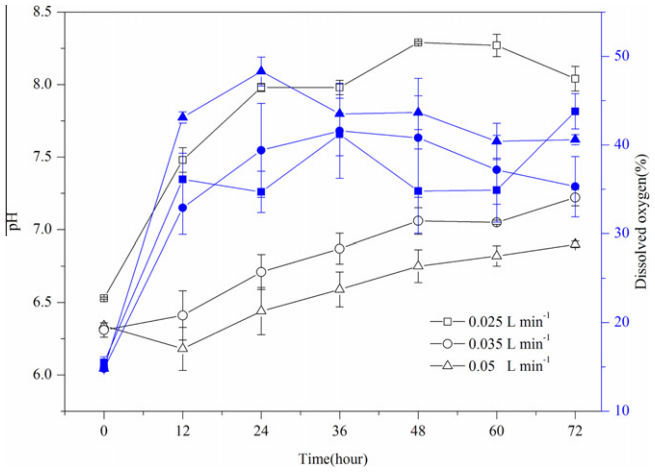

Fig. 4. The pH values and DO levels of culture in CRWP. Open and closed symbols represent pH values and DO levels of culture, respectively.

and calculated values of dissolution efficiency was 14%, which showed a goodness of fit of the equation. As shown in Eq. (4), the efficiency of CO_2 dissolution in the CRWP aerated with certain CO_2 concentration had no relation with gas sparging rate and may be determined by the characteristics of CRWP, including the structure of photobioreactor, liquid velocity, mixing time and temperature, which influenced the overall volumetric mass transfer coefficient and hydrodynamics characteristics (Ugwu et al., 2008). The maximal CO_2 dissolution efficiency of media in CRWP aerated with 15% CO_2 was 64% according to Eq. (4), which was the maximal CO_2 fixation efficiency by microalgae under continuous CO_2 supply because microalgae can only utilize dissolved CO_2 and change it into biomass.

3.2. Microalgal productivity and CO_2 fixation efficiency with continuous gas sparging

The microalgal productivity and CO_2 fixation efficiency in CRWP under the concentration of 15% CO_2 with different gas flow rates are shown in Table 1. The changes of pH values and DO levels of culture during microalgal growth are shown in Fig. 4. As shown in Fig. 4, for 0.035 and 0.05 L min^{-1}, the pH values of culture ranged from 6.18 ± 0.14 to 7.22 ± 0.05, which did not inhibit microalgal growth (Hodaifa et al., 2010), and the levels of DO varied from 14.8 ± 0.1% to 48.3 ± 1.6% during cultivation. Although the oxygen build-up in photobioreactors would lead to photorespiration and even photo-oxidation with high levels of irradiance (Douskova et al., 2009; Miron et al., 1999), the competitive inhibition of ribulose-1,5-bisphosphate carboxylase by oxygen gradually became weak with the increase of concentration of CO_2 when oxygen level ranged from 20% to 50%(Aiba, 1982). In the present study, because the microalgal culture was aerated with high concentration of CO_2 (15% CO_2), and illuminated with low light intensity (100 μmol m^{-2} s^{-1}), the inhibition action of oxygen accumulation to microalgal growth in the CRWP did not be found. Therefore, the culture pH and DO level in CRWP did not inhibit microalgal growth with the gas sparging rates of 0.035 and 0.05 L min^{-1}. Fig. 5 gives the change of dissolved CO_2 concentration in culture aerated with different gas sparging rates. As shown in Fig. 5, the average dissolved CO_2 concentrations in microalgal culture aerated with the gas sparging rates of 0.025, 0.035 and 0.05 L min^{-1} from 12 to 72 h were 73, 713 and 1441 μmol L^{-1}, respectively. The microalgal productivity in CRWP aerated with the gas sparging rate of 0.025 L min^{-1} was obviously lower than that of other gas flow rates, which showed that the dissolved CO_2 amounts cannot satisfy microalgal growth. Weissman et al. (1988) observed that CO_2 concentration in bulk liquid of at least 65 μmol L^{-1} and pH 8.5 were required for optimal productivity of some microalgae, but in the present study, CO_2 concentration of 73 μmol L^{-1} in media and pH 8 (shown in Fig. 4) cannot meet microalgal normal growth. The difference may be determined by algal species. For 0.035 and 0.05 L min^{-1}, with the increase of gas flow rate, the concentration of dissolved CO_2 in culture increased and the CO_2 fixation efficiency decreased, which indicated that much of CO_2 supply was wasted with continuous supply. According to Eq. (4), the maximal microalgal productivity in CRWP aerated with gas flow rates of 0.035 and 0.05 L min^{-1} should be 0.3970 and 0.5670 g d^{-1} L^{-1}, respectively, but the experimental microalgal productivity was 0.3496 and 0.3219 g d^{-1} L^{-1}, respectively, which also indicated that some of supplied CO_2 was wasted.

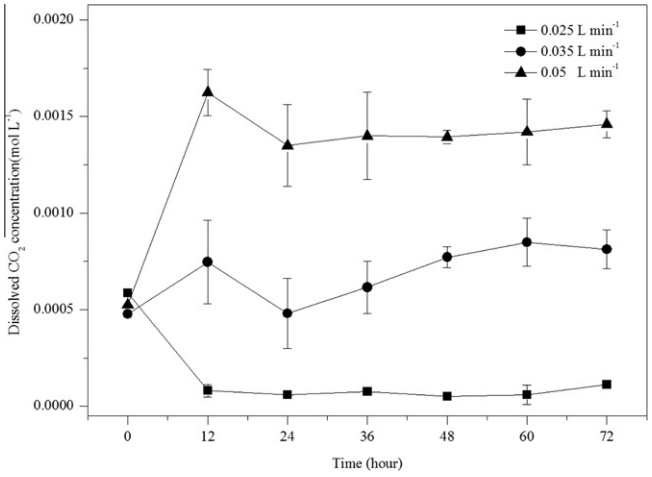

Fig. 5. Dissolved CO_2 concentrations in culture with different gas sparging rates.

3.3. CO_2 fixation efficiency by microalgae with intermittent gas sparging

With continuous gas sparging rates of 0.035 and 0.05 L min^{-1}, the fixation efficiency was 56% and 36%, respectively. According to Eq. (4), even the dissolved CO_2 was completely fixed by microalgae, the maximal CO_2 fixation efficiency in CRWP aerated with 15% CO_2 was 64%, which indicated there were much CO_2 escaped from CRWP. The method to further improve CO_2 fixation efficiency in CRWP was to increase the length of raceway, which would prolong the residence time of CO_2 in CRWP, or supply CO_2 intermittently. In this work, the later was further studied. In order to reach the maximal CO_2 fixation efficiency of 100%, the amounts of supplied CO_2 should completely dissolve in culture, and meanwhile, the dissolved CO_2 met microalgal growth' need. Therefore, the relation between the gas sparging time (t_{aer}), the stopping time (t_{sto}), CO_2 dissolution rate in gas sparging (v_{dis}) and stopping (v_{res})

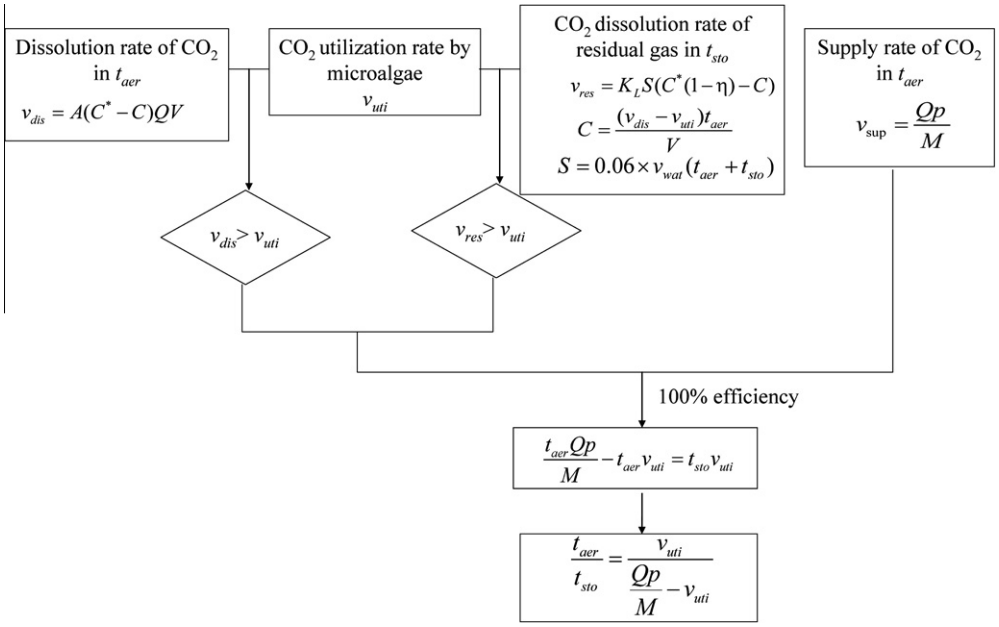

Fig. 6. The calculation model of intermittent time (0.06 is the width of each channel in the closed raceway pond).

time and the rate of CO_2 utilization (v_{uti}) by microalgae should be studied. In the stopping time, the remaining CO_2 (including CO_2 in culture and residual gas) should be enough for microalgal growth, and CO_2 in residual gas could dissolve completely in culture in t_{sto}. The dissolution rate of CO_2 in residual gas (v_{res}) can be expressed as follows:

$$v_{res} = K_L S(C^*(1-\eta) - C),$$

where K_L is equal to 0.0096 m min^{-1} (Shah et al., 1982) and S is the surface area of residual gas bubble in CRWP.

In the gas sparging time, CO_2 dissolution rate (v_{dis}) can be calculated from Eq. (3), and the average microalgal productivity (P) can be used to calculate the rate of CO_2 utilization (v_{uti}) by microalgae. Fig. 6 gives the calculation model of intermittent time. For example, under the gas flow rate of 0.035 L min^{-1} with 15% CO_2 at 24 °C, the average microalgal productivity was 0.3496 g d^{-1} L^{-1} with continuous gas sparging (shown in Table 1). Through calculating, the dissolution rate of CO_2 (v_{dis}) was greater than the utilization rate of CO_2 by microalgae (v_{uti}) in the gas sparging time, and in the stopping time, v_{res} was far greater than v_{uti}, which indicated that the utilization rate of CO_2 by microalgae was the rate-limiting step for CO_2 absorption and the stopping time (t_{sto}) was determined by the utilization rate of CO_2 by microalgae (v_{uti}) in the intermittent operation. Therefore, the amounts of CO_2 supply in the gas sparging time should be equal to that of CO_2 of microalgal utilization in the gas sparging and stopping time, which gives:

$$\frac{t_{aer}Qp}{M} = t_{aer}v_{uti} + t_{sto}v_{uti};$$

For the gas sparging rate of 0.035 L min^{-1} in CRWP, the proportion of t_{aer} to t_{sto} was closed to 1. The batch microalgal culture with the gas sparging time of 10 s and the stopping time of 10 s was investigated. After five days of cultivation, the average microalgal productivity was 0.2935 ± 0.0023 g d^{-1} L^{-1}, which indicated that the CO_2 fixation efficiency by microalgae reached 95% in CRWP with intermittent gas sparging. Compared with continuous supply of CO_2, the intermittent gas sparging decreased the amounts of CO_2 supply and meanwhile the amounts met microalgal growth' need, which improved the fixation efficiency. The values of t_{aer} and t_{sto} should be further analyzed. Under intermittent gas sparging, with the utilization of CO_2 by microalgae, the supplied CO_2 should completely dissolve in culture before running out of the CRWP, which meant that t_{aer} should be shorter than the residence time of CO_2 in CRWP with continuous supply, and on the other hand, the shorter of t_{aer} and t_{sto}, the faster of dissolution rate of CO_2 in culture, which benefitted the CO_2 fixation by microalgae. However, the microalgal productivity should be determined previously. Microalgal productivity can be predicted depending on light transfer with rich nutrition in medium (Fouchard et al., 2009; Yun and Park, 2003). Through the relationship between microalgal productivity, light distribution and CO_2 dissolution rate, the proportion of t_{aer} to t_{sto} can be calculated.

4. Conclusions

In this work, the CO_2 mass transfer, dissolution efficiency and CO_2 fixation efficiency by microalgae in the closed raceway pond were investigated. The dynamic equation for CO_2 dissolution and the method of improving CO_2 fixation efficiency by microalgae were developed. The experimental results showed that the developed model provided the guide for the improvement of CO_2 fixation efficiency by microalgae. Through the developed model, under intermittent operation (10 s:10 s), the fixation efficiency reached 95%. The concrete time of intermittent operation with different CO_2 partial pressures and gas flow rates should be further studied.

Acknowledgements

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