Jurnal Teknologi

Full paper

Preliminary Study on Biomitigation Green House Gas Carbon Dioxide in Closed System Bubble Photobioreactor: Relationship Among the Mass Transfer Rate and CO_2 Removal Efficiency in High Level of CO_2

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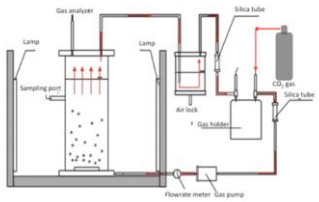
Article history

Received :1 January 2014
Received in revised form :
15 February 2014
Accepted :18 March 2014

Graphical abstract



Abstract

Emission of carbon dioxide (CO_2) is a major contributor to global warming. Biofixation of CO_2 by microalgae in photobioreactors seems to be a promising strategy for CO_2 mitigation. The research to determine the overall mass transfer coefficient (kLa) has been done to find the way on biomitigation CO_2 emission by using biologically Carbon Capture and Sequestration method. This research was conducted according to green microalgae *Scenedesmus obliquus* activity, which is cultivated in a bubble photobioreactor through the mass transfer process that assumed adequate mixing occurs. Flow rate of CO_2 that supplied to the system were 2 L/min, 5 L/min and 8 L/min, when each rate flowed into the photobioreactor with high CO_2 concentration (v/v) of 2%, 5% and 10%. The highest CO_2 removal efficiency occurred at culture that supplied with an CO_2-enriched air flow rate of 5 L/min. The kLa (CO_2) value is the highest in 0.3582/day at 2% CO_2 concentration and flow rate of 2 L/min, while the lowest is in 0.0503/day at 5% CO_2 concentration and flow rate of 8 L/min. In terms of solubility is inversely proportional to the flow rate, the less carbon dioxide is dissolved at the rate of 8 L/min as well as the value of the kLa. The results showed that the variation of flow rate will affect the amount of mass transfer coefficient, growth rate and cell biomass. Higher flow rate decreases kLa value as well as CO_2 removal efficiency.

Keywords: Biomitigation; photobioreactor; k_La (CO_2); mass transfer; *Scenedesmus obliquus*

1.0 INTRODUCTION

Removal of carbon dioxide (CO_2) from industrial flue gases become an urgent necessity in order to reduce the impact of CO_2 on global warming. The researcher [1] explained that the two main CO_2 mitigation strategies commonly used approach is based on chemical reactions and biological mitigation. On the one hand, the chemical reactions based CO_2 mitigation approach is energy-consuming, expensive process to use, and disposal problems due to either carbon dioxide captured and absorbent waste needs to be removed. On the other hand, one interesting method for CO_2 mitigation is the use of biological processes in the utilization of CO_2 directly from the biomass to be converted into a point source of CO_2 emissions in a closed system that is engineered as a photobioreactor. The biological CO_2 mitigation has attracted much attention in recent years because it causes the production of biomass energy in the process of fixation of CO_2 through photosynthesis. Other researcher [2] described the use of photobioreactors for CO_2 absorption by microalgae offers major advantages is the increase in productivity of microalgae for controlled environmental conditions and maintenance space utilization can be optimized so that it is considered more efficient than using terrestrial plants that require extensive and expensive land.

The fact that most of the flue gases produced by most industries contain 5–15% CO_2 concentration gives microalgae the advantage of being the best candidate for creating a sustainable carbon sink. One of the hydrodynamic variables related to the growth of microalgae, and the effectiveness of the removal of carbon dioxide is the mass transfer of carbon dioxide. The mass transfer of carbon dioxide from air into the media can be growth- limiting in dense algal cultures [3].

In 2009, [4] stated that the transfer of CO_2 from a gas to a liquid depends on many parameters such as gas flow rate, CO_2 partial pressure, bubble diameter and lifetime can have large influences on the rate of transfer. Mass transfer coefficient of CO_2 or k_La (CO_2) could reflect the condition of the mass transfer that occurs in the photoreactor. The k_La term generally used to describe the overall volumetric mass transfer coefficient photobioreactor. Volumetric mass transfer coefficient (k_La) is characteristic of a photobioreactor in determining the ability of a photobioreactor to maintain optimal cell growth. The k_La value (CO_2) is a general hydrodynamic parameter that is often used to assess the performance of the bioreactor in the cultivation of microalgae. For microalgae cultivation process is necessary k_La

optimal value. The high value of k_La indicates the mass transfer of CO_2 better in microalgae culture [3], [4], [5], [6].

Behavior of k_La and cell growth rate varies in different regions in a fluid stream. Region fluid flow in the photobioreactor can be divided into streams of bubbles, and heterogeneous transition zone depends on the gas velocity, the flow area of the bubble, the gas hold-up, the interface area and the k_La proportional to the superficial gas velocity [5]. Despite the reduction in interface area start moving from the transition zone to zone heterogeneous but the gas continues to rise and reaches a high value of the k_La. The k_La values, a specific increase in the initial growth but at the end of the transition zone begins to decline [7].

This study examines the effect of the concentration of carbon dioxide on CO_2 removal by microalgae *Scenedesmus obliquus*, biomass concentration, and the rate of mass transfer in a bubbles photobioreactor.

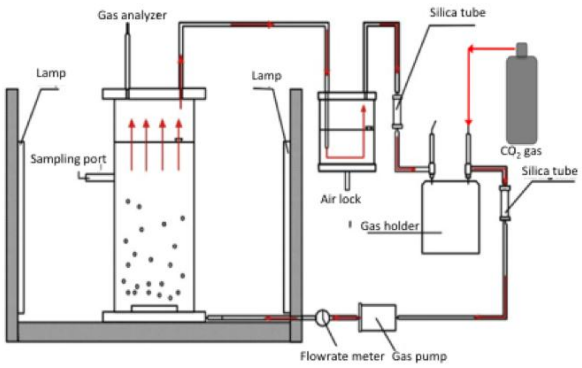


Figure 1 Schematic design of closed system bubble photobioreactor microalgae

2.0 MATERIAL AND METHODS

2.1 Culture of *Scenedesmus obliquus*

Microalgae used in this study is green microalgae *Scenedesmus obliquus* isolated from Wastewater Treatment Bojongsoang, Bandung, Indonesia previously been performed screening of the types of microalgae found in the installation of a pool [8]. Provasoli Hematococcus Media (PHM) chosen as growth medium because the medium is suitable for freshwater microalgae, such as *Scenedesmus obliquus* [9]. In the activation process, *Scenedesmus obliquus* put in PHM medium in glass bottles, aerated with air at a rate of 800 ml/min, at 2500 lux light intensity and ambient temperature. The activation process is a growing process of microalgae cells to be used directly in the photobioreactor cultivation. This process also serves to prevent long a lag phase in the photobioreactor cultivation.

2.2 Operational Experimental Condition of Closed System Photobioreactor

This study used a vertical type with a bubbles photobioreactor to distribute CO_2-enriched air (Figure 1A) whereas sparger placed at the bottom of the photobioreactor to distribute carbon dioxide (Figure 1B). One of the parameters that determine the growth and biofixation CO_2 by microalgae is the rate of mass transfer in a photobioreactor gas. In order to enhance the mass transfer, a Sparger as gas converter into small bubbles is fitted at the bottom of the reactor . Sparger used has a diameter of between 100-160 µm. Sparger is mixing condensed gas with a flow rate of 2 L/min, 5 L/min and 8 L/min to increase the mass transfer of CO_2 and also eliminates the O_2 produced during photosynthesis. The size of the bubbles are formed also affect CO_2 fixation by microalgae.

Advantage of using the photobioreactor microalgae cultivation process is providing culturing conditions that can avoid contamination as well as presence of competitors or culture in a state that has been adjusted in order to obtain the optimum results for then can be done to scaled up. Environmental conditions for the study were light intensity of 4000 lux, the periodicity of 16 light hour and 8 dark hour and temperature of 30^OC.

2.3 Measurement of Biomass Concentration and Growth Rate of *Scenedesmus obliquus*

Biomass concentration (dry weight) according to [10] calculated by the formula: dry weight (X; mg) = y (mg) - x (mg). Specific growth rate (μ; d^{-1}) was calculated as follows:

$$\mu = \frac{1}{X}.\frac{dX}{dt} \quad (1)$$

Dry weight cell biomass of microalgae was obtained by evaporating the liquid in the cell culture. A total of 100 mL culture tube inserted into centrifuges, and then centrifuged at 3500 rpm for 10 minutes [11]. Supernatant was then removed from the tube pasta until just earned cells. Pasta cells were then put into a petri dish that had previously been weighed (x). Samples were put in the oven with a temperature of 105^0C for one night to get a constant weight (y), and then stored in a desiccator for 30 minutes before re-weighed.

2.4 Measurement of CO_2 Concentration and Determination of CO_2 Removal Efficiency

Efficiency of CO_2 removal can be calculated by the following formula:

$$\frac{\text{Influent of } CO_2 - \text{Effluent of } CO_2}{\text{Influent of } CO_2} \times 100\% \quad (2)$$

The CO_2 concentration in the influent gas and effluent gas was measured by Portable Combination Gas Detector RIKEN Model RX-515.

2.5 Measurement of Overall Mass Transfer Rate

The rate of CO_2 absorption associated with the mass transfer of CO_2 and the overall rate of carbon dioxide by microalgae biofixation. CO_2 absorption rate is temperature dependent. In the overall mass transfer rate equations contained in the Equation 3.

$$Na = k_L a\,(C^* - Ct) \qquad (3)$$

Na is the rate of transfer of carbon dioxide per unit of time. The equation shows that the rate of transfer of carbon dioxide per unit time is proportional to the difference between the concentration of the injected carbon dioxide saturated with dissolved carbon dioxide concentration. In accordance with the theory of the reaction order and reaction rate, the rate of mass transfer of CO_2 is included in the reaction order 0 [3], [5].

■3.0 RESULTS AND DISCUSSION

3.1 CO_2 Removal Efficiency

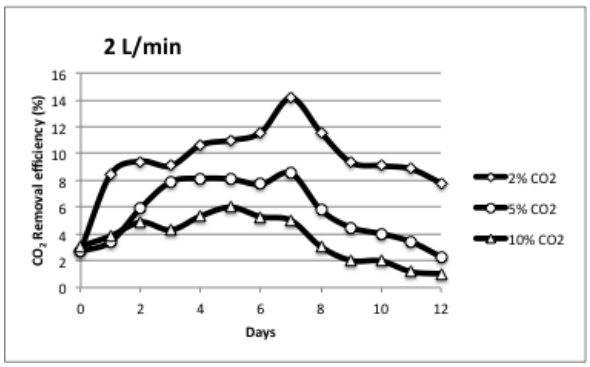
Figure 2 shows the profile of CO2 removal efficiency at various flow rates during 12 days of observation. At each slow flow rate (2 L/min) , moderate (5 L/min), and rapid (8 L/min), the CO_2 removal efficiency is higher in cultures were flowed with 2% CO2, followed by culture that flowed with 5% CO2 and 8% of CO_2. Low flow rates allow CO_2 gas stuck longer in the system, while the rapid flow rate causes turbulence, and CO_2 is rapidly moving out of the system so that CO_2 removal could not occur. In other words, the longer of detention time at a lower flow rate (2 L/min) causes the higher of CO_2 removal efficiency.

Although high CO2 removal efficiency generally occurs in cultures were flowed with a slow flow rate (2L/min) but the highest removal efficiency of the CO_2 occurred in the reactor with a flow rate of 8 L/min i.e the CO_2 removal efficiency was 16% in cultures supplied with 2% CO_2 (Figure 2c). The researcher [10] reported that 24% CO_2 has been sequestered in microalgae cultures that supplied with 5.5% pure CO_2. Thus the efficiency of CO_2 removal in this study is low, although the efficiency is still greater than the volume of carbon dioxide in the ambient air.

Average of CO_2 removal efficiency at various flow rates can be seen in Figure 3. For every input of CO_2 concentration with variations of flow rate, has the same efficiency profile, whilst the higher the concentration of CO_2 the smaller the average removal efficiency of CO_2

Reduced amount of CO_2 that captured in the medium of microalgae in a photobioreactor will be used for the photosynthesis process by microalgae. CO_2-enriched air that flowed into the photobioreactor is a potential gas to perform mass transfer. In hydrodynamic aspect, its purpose is to ensure the more gas hold in the system then the mass transfer process will be optimal

In the water, the carbon dioxide will dissolved and formed a carbonate compound depends on the pH conditions. Microalgae will use the carbon dioxide dissolved in water to perform photosynthesis process and store it in the vacuole. Furthermore, by using Rubisco enzymes used in photosynthesis process which will naturally produce oxygen, so that the carbon dioxide out of the photobioreactor will be less than the amount that supplied.



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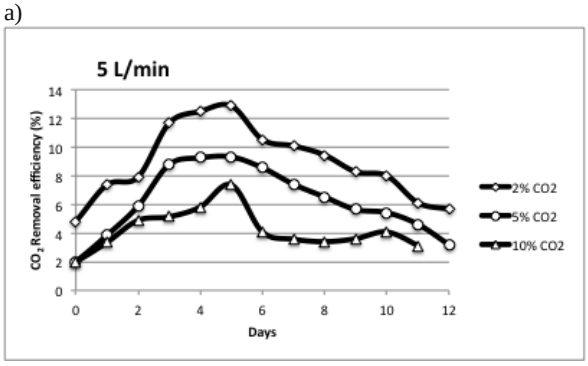

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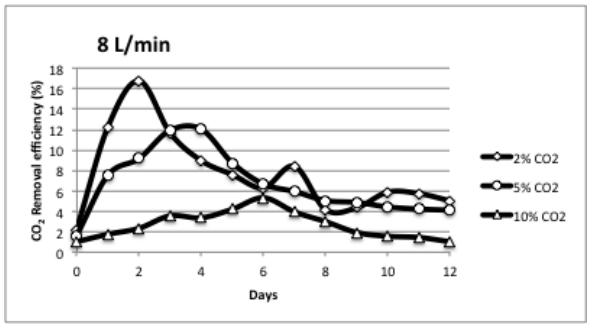


c)

Figure 2 Profile of CO_2 removal efficiency with high level concentration of CO_2 at variation of flow rate a) 2 L/min, b) 5 L/min, c) 8 L/min

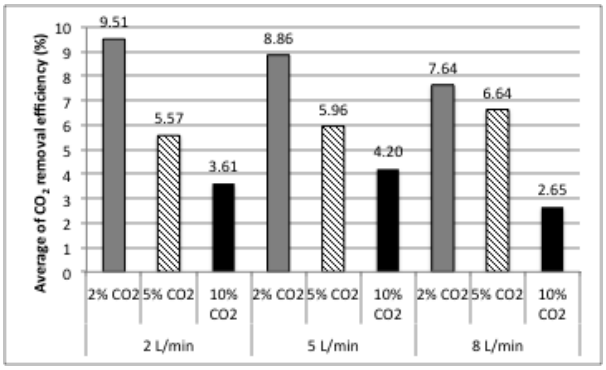


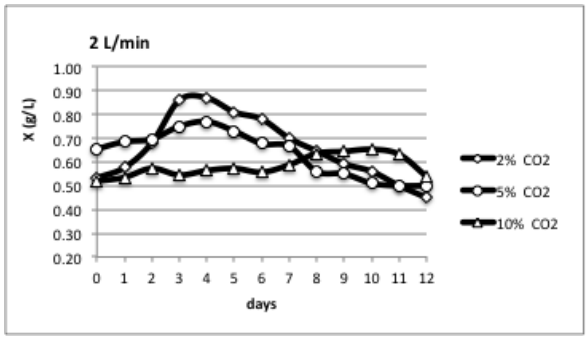
Figure 3 Average of CO_2 removal efficiency with variation flow rate and concentration (v/v) of CO_2

3.2 Organic Biomass of *Scenedesmus obliquus*

According to research by [12], microalgae *Scenedesmus obliquus* is appropriate to reduce the concentration of CO_2 for biomass

productivity and high lipid and carbon fixation ability is higher than other microalgae. Or fixation of CO_2 removal efficiency depends on the physiological condition of microalgae, such as the potential for cell growth and metabolic capabilities of CO_2.

Microalgae growth can be seen in Figure 4. In the cultures supplied with CO_2 of 2 L/min, growth tends to be more stable than other flow rates. Microalgae growth rate increased until at the height of growth, then the growth rate slows. At each flow rate variation, the same phenomenon occurs but in different days, such that the flow rate becomes a factor that limits the growth of microalgae.



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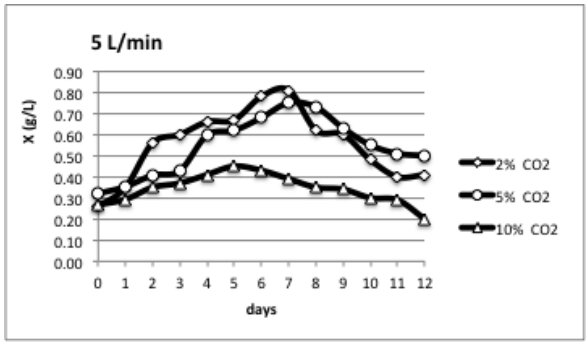

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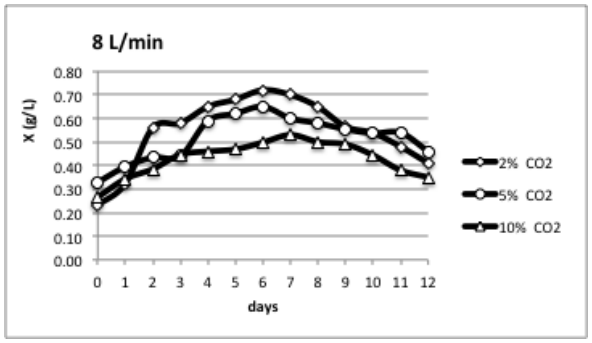

c)

Figure 4 Profiles of organic biomass concentration (g/L) of *Scenedesmus obliquus*

Figure 4 shows the growth response of microalgae *Scenedesmus obliquus* at elevated flow rate and concentration (v/v) of CO_2. These profiles indicate that the dry weight of biomass as growth response is affected by the flow rate and concentration of CO_2 that injected into photobioreactor. In each culture were be flowed with 10% CO_2, at condition a slow flow rate (2 L/min) to fast flow rate (2 L/min), all cultures showed poor growth response compared with cultures were be flowed with a lower CO_2 concentration than 10%, i.e 2% and 5% CO_2. The average biomass concentration in culture were fed with the 10% CO_2 was about 30% compared with an average biomass obtained from cultures were fed with 2% CO_2 and 5% CO_2 (Figure 5).

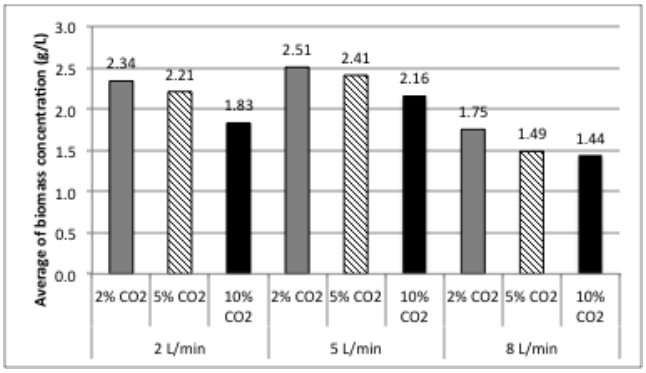

Figure 5 Average of biomass concentration(%) with variation flow rate and concentration (v/v) of CO_2

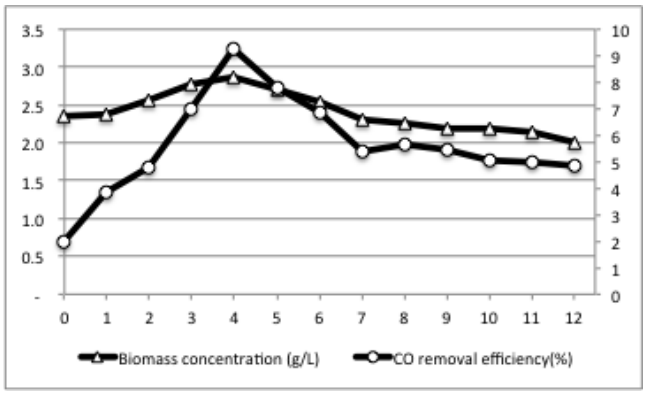

Figure 6 Profiles of CO_2 removal efficiency and concentration of biomass at condition of 5% (v/v) CO_2 and Flow rate of 5 L/min

In this study, *Scenedesmus obliquus* still could grow at concentration of 10% CO_2, with flow rate of 8 L/min or 27.16 m/hour although it is potentially cause the shear stress. The shear stress occurs when superficial velocity exceeds the ability of microalgae to withstand the stress.

Microalgae growth in the medium supplied with carbon dioxide-enriched air will be hampered due to lack of other substrates than carbon dioxide. In this study, CO_2-enriched air is the only addition compounds that injected into the system. CO_2 removal efficiency at flow rate of 2 L/min did not differ significantly with culture were be flowed with flow rate of CO_2 5 L/min for each concentration of CO_2 is put into the system (Figure 3) but we recommend the flow rate of 5 L/min as the best flow rate, not too slow (2 L/min) nor too fast (8 L/min), since generally the maximum biomass concentration (g/L) of all elevated CO_2 level occurred in cultures supplied with CO_2 flow rate of 5 L/min. When compared with the findings of [15], the maximum cell concentration of 2.02 g/L was found in cultures supplied with 5% CO_2 and a minimum cell concentration of 1.16 g/L was found in cultures supplied with 0.5% CO_2. It is therefore advisable to maintain the concentration of CO_2 is lower than 5% because higher CO_2 concentrations can inhibit the growth of microalgae. In addition, they reported a gradual increase in the concentration of the cells when the concentration of CO_2 increases. Therefore,

the concentration of CO_2 is directly related to the cells concentration and microalgae biomass.

Related to the growth of microalgae, the concentration of carbon dioxide in the culture medium could not below the requirement for maximum growth rate culture but should not exceed the maximum that can be tolerated by a particular microalgae. In comparison with the study of [14], 5% to 20% concentration of CO_2 from flue gas is the optimum carbon source that is beneficial to the growth of biomass. At higher CO_2 concentrations of 20% will cause the pH and carbonate anhydrase activity is reduced, so that will inhibit growth. The researcher [14] also concluded that the microalgae growing well below 5% to 20% of CO_2, despite the best growth potential at 10% CO_2.

In this study, microalgae could grow in culture that injected 10% CO_2 concentration with a flow rate of 8 L/ min, although the growth rate tends to be stable during this phase according to a constant number of cells. It can be caused by reduction in the nutrient medium or by the accumulated toxic products of metabolism resulting in impaired growth. In most cases, cell turnover occurs in the stationary phase, the cell loss occurs slowly because death s balanced by the formation of new cells by division. If this is the case then the number of cells will increase slowly even though the number of living cells remained constant. The optimal growth can be determined by calculating the microalgae growth rate (μ) in the exponential phase. The fastest growth rate occurs in the cultures supplied with CO_2 flow rate of 5 L/min, which is the highest growth rate achieved by microalgae at 0.34 day^{-1} for 8 L/min flow rate and concentration of 2% CO_2, when compared with the experiment was done by [13] also exploited *Scenedesmus obliquus* at a specific growth rate of 0.44 day^{-1}, then the result is not much different.

Comparison of microalgae growth rate is influenced by the design of the photobioreactor that used, the flow rate is high with larger bubbles and high superficial velocity that can inhibit growth, in addition to the optimum conditions can accelerate the mass transfer, so that this value will be compared with the rate of overall mass transfer.

CO_2 transfer efficiency is one of the most important parameters to improve the level of CO_2 removal efficiency and biofixation by microalgae in a photobioreactor culture system. To determine the relationship between the biomass concentration by dry weight and removal efficiency of CO_2, linked to the purpose of this study, the data used flow rate of 5 L/min and 5% CO_2 (Figure 6). The CO_2 removal efficiency increase as well as biomass concentration, on the contrary when the CO_2 removal efficiency decrease, the biomass concentration will decrease also. CO_2 absorption by microalgae cells allegedly stimulated by an increase in dissolved of CO_2 in the culture media, so that the culture without the addition of CO_2 will make absorption rate of CO_2 is low.

This is consistent with [13] who described when CO_2 added to the growth medium, the growth rate of the photosynthetic microorganism will be increase, otherwise when there is no CO_2 is added to the culture then there is no growth rate. Described in the experiment that uses microalgae *Scenedesmus obliquus* and *Spirulina Sp.*, microalgae can be grown in a growth medium with a given CO_2-enriched air. This indicates that *Scenedesmus obliquus* can fix CO_2, and so that CO_2 is a limiting factor of growth of *Scenedesmus obliquus*.

High CO_2 concentration in the closed system proved to be effective to increase the growth rate, using the CO_2 bubbles as the transfer medium, rather than doing the same thing in the ambient air. However, the growth rate drops when the culture is conducted with very high concentrations and flow rates of CO_2. Extreme high flow rate of CO_2 can increase carboxylation microalgae and suppress the activity of rubisco oxygenation, thus increasing photosynthesis.

3.3 Carbon Dioxide Mass Transfer Coefficient

Mass transfer of carbon dioxide in the photobioreactor will also be influenced by the hydrodynamic aspects, one of which is the superficial velocity of the gas in the photobioreactor. Superficial velocity (Ug) also called volumetric flux, the gas volumetric flow rate divided by the cross - sectional area of the fermenter [16]. Photobioreactor diameter used is 15 cm, then the surface area of the tube cross section 1.767 x 10^{-2} m^2, while the injected air flow rate of 2 L/min, 5 L/min and 8 L/min so that the value obtained using Equation 3.3 superficial velocity for each flow rate of 1.9 x 10^{-3} m/s, 4.7 x 10^{-3} m/s and 7.5 x 10^{-3} m/s (Table 1).

Table 1 Variation of flow rate of carbon dioxide and superficial velocity

Flow rate (L/min)	Surface are of photobioreactor (m^2)	Superficial velocity (m/s)	Superficial velocity (m/hour)
2	1,767 x 10^{-2}	1,9 x 10^{-3}	6,76
5	1,767 x 10^{-2}	4,7 x 10^{-3}	16,98
8	1,767 x 10^{-2}	7,5 x 10^{-3}	27,16

In Indonesia, [17] describes the use of both superficial gas flow rate which indicates ideal mixing is between 1.2 < Ug < 12 m/hour. When referring to this, the flow rate of 2 L/min was included in the ideal mixing, while the flow rate of 5 L/min and 8 L/min excluding superficial velocity ideal for mixing, but this will be assessed by observing the growth of microalgae, the rate of transfer mass and carbon dioxide fixation by microalgae.

Mass transfer of carbon dioxide is affected by mass transfer rate of carbon dioxide. Determination of carbon dioxide transfer rate is very important given the close relationship between biological and physicochemical processes and the concentration of CO_2 in solution. Low pH values and high aeration conditions led to a decrease in available inorganic carbon as CO_2 stripping. A study shows the importance of pH on inorganic carbon limitation for algae biofilm growth. Conversely, when the level of the lower CO2 stripping, for example, using a low agitation and / or aerated culture conditions, the concentration of dissolved CO_2 will rise above the saturation value (saturation concentration) [18].

The rate of mass transfer coefficient is determined by the concentration of carbon dioxide saturation that is injected CO_2 concentration and calculated concentration in water using Henry 's constant, which Henry constant gas into a liquid that is used is from 10 to 1.5 $mol.L^{-1}.atm^{-1}$ [19]. When there is a mass transfer of carbon dioxide from the gas phase to the liquid phase, only dissolved CO_2 of all species of CO_2 is responsible for the mass transfer of carbon dioxide through the gas-liquid interface [3].

The concentration of dissolved carbon dioxide is obtained by using the calculation of acidity and alkalinity, and put it into the Equation 2.21. The results were plotted on a graph between ln $[(C^*-C_0)/(C^*-Ct)]$ at intervals (days), and the gradient of the curve is a line of mass-transfer coefficient (Figure 7).

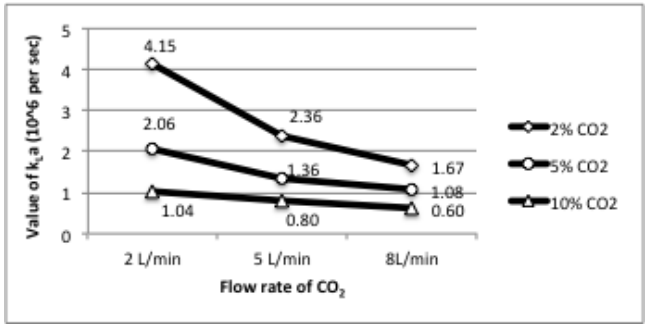

Figure 7 The k_La values in variation of flow rate of CO_2

The k_La highest value of 4.15 x 10^6 sec^{-1} occurs in cultures were fed with 2% CO_2 with a flow rate of 2 L/min. With a flow rate of 2 L/min then the diameter of the bubbles that form tend to be small and the detention time within the photobioreactor to be longer. Figure 7 describes any increase in the flow rate will decrease causing the value of the k_La. Similarly, at the same flow rate then any increase in concentration led to a decrease in the value of the k_La, except at a flow rate of 8 L/min. The k_La values generally increased with increasing flow rate of the fluid tangential, because the depletion layer liquid limit .

The k_La value of this study are much smaller when compared to research of [6], which uses bubbling CO_2 obtained k_La value 7.0 x 10^{-3} min^{-1}. In another experiment conducted [17], using a reactor volume of 18 L and 40 L, a combination of a flat plate and bubble columns, the k_La values obtained for 0.303 min^{-1} or 0.005 sec^{-1}. In other studies, for the bubble column reactor as compared to other types of reactors, has a small value so that the k_La can be concluded that although the bubble column reactor is simpler and easier in operation, but provides little value of k_La. Photobioreactor design seems to have impacted the ability of mass transfer.

3.4 Relationship Among the Mass Transfer Rate, CO_2 Removal Efficiency and Biomass Concentration

Cultivation process of microalgae in closed system require optimal value of kLa because this value indicate a high CO_2 mass transfer process well occurred in microalgae culture, but the value of kLa is not always a good one for the growth of microalgae culture. The researcher [20] stated that the values of kLa that too high may lead to the occurrence of shear stress on the microalgae which could inhibit the growth of microalgae, which cause the biomass content will be reduced, so that should be avoided.

Mass transfer of CO_2 from the air into the growth medium may limit the growth of microalgae. By comparing the growth rates of microalgae and mass transfer, it shows that performance is highly dependent on the bioproduction of microalgae metabolism and magnitude carried microalgae growth rate is influenced by the mass transfer of CO_2 into the cell which would then be used for the sustainability of photosynthetic activity by microalgae. In this study the profile of coefficient k_La inversely with the growth rate of microalgae. The higher the flow rate at each increment, the smaller the value of microalgae growth rate, thus the smaller the mass transfer. It occurs in cultures fed by 5 L/min of CO_2. The magnitude of the growth rate is affected by the mass transfer of CO_2 into the cell which would be used for the sustainability of microalgae photosynthetic activity.

Removal of CO2 in liquid phase is affected by the volumetric mass transfer coefficient and affects the rate of growth of microalgae biomass. This happens on a large flow rate with the highest superficial velocity, thus indicating ideal mixing. In other case, [5] explained that the increase in the rate of mass transfer of CO_2 has been described as a result of natural convection, the process of destabilization of the gas-liquid boundary layer caused by the increase viscosity/specific gravity (density) of dissolved CO_2 after CO_2 dissolved in water. In other words, the driving force on natural convection occur when the water with CO_2moves upward toward the boundary layer (interface) and lower CO_2 viscosity of water moving downward, so as accelerate the mass transfer rate. In addition, changes in the density of the liquid CO_2 is highly dependent on the concentration of CO_2 (initial pressure of the gas) i.e, the higher concentration will make the CO_2 to increasingly dissolve in the water.

■4.0 CONCLUSION

The relation between CO_2 that flowed into the system, CO_2 solubility in the growth medium and the rate of mass transfer coefficient has been described. CO_2 removal efficiency is affected by flow rate and concentration of CO_2. The results showed that the flow rate significantly affects the operational conditions of photobioreactor that are observed through microalgae growth rate, biomass concentration of cells and the rate of mass transfer. CO_2 flow rate that supplied to the system is inversely proportional to the mass transfer coefficient. Flow rate is too high (8L/min) causing turbulence that causes sheer stress on the cells and CO_2 detention time in the system did not last long. This condition also gives the value of the mass transfer coefficient so low that the inorganic carbon cannot be fully utilized for the growth of *Scenedesmus obliquus*.

Acknowledgement

We would like to thank DIKTI (Directorate General of Higher Education Indonesia) Programme of Decentralization 2014 for funding some of this research.

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