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Design of an Energy-Saving Membrane Separation Module for Algae Cultivation

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In this study, focusing on a zeolite membrane separation method expected energy-saving, appropriate environments for algae cultivation were investigated from the CO_2 capture aspects. In addition, the authors elucidated the membrane separation performance and its energy consumption through experiments and simulations for the integration of CO_2-treated processes. The results suggested that only 2-15 % CO_2 in air ventilation was required for several algae growth. To satisfy the CO_2 concentration constraint, the membrane could separate CO_2 to approximately 15 % with the energy required for 0.76 MJ/kg-CO_2 and a recovery ratio of 71 %. The combination of membrane separation and algae cultivation from low concentration CO_2 sources is a promising method for carbon capture and utilisation.

1. Introduction

Carbon Capture and Storage (CCS) and Carbon Capture and Utilisation (CCU) to suppress global warming have been attracting attention because carbon dioxide (CO_2) causes global warming as a greenhouse gas. On the contrary, CO_2 is an essential material for refrigerant (dry ice), carbonated drinks, laparoscopic surgery (Ilkben et al, 2021), welding as sealing gas (Math et al., 2021). Recently, large and high concentration CO_2 sources such as steel-works have been shutting down gradually due to world economics and environmental protections. Thus, it is required that CO_2 emitted from low concentration CO_2 sources such as thermal power generation plants or garbage incineration plants is separated to protect environments and to satisfy a variety of CO_2 usage.

There have been some methods such as chemical absorption, physical adsorption, membrane, and cryogenic separation methods to separate CO_2 from a gaseous stream. The most commonly used method for CO_2 capture in industries among the above-mentioned methods is a chemical absorption method with amine solutions (Teranishi et al., 2016). In this method, CO_2 reacts chemically with amine solutions. Therefore, this process inherently produces pure CO_2 and achieves a high CO_2 recovery ratio. However, it is well-known that the process is energy intensive (Goto et al., 2009). Thus, energy-saving technology has been required and investigated (Goto et al., 2016). Nagumo et al. (2013) reported that membrane separation required the least energy-required method among the other CO_2 capture methods. In addition, Basile et al. (2010) reported that a membrane separation method is an alternative solution for separating CO_2 from off-gas of an integrated gasification combined cycle (IGCC).

Some researchers have investigated several CO_2 utilisation methods such as artificial photosynthesis to chemicals (Jia-Way et al., 2018), biological digestion (Alami et al., 2021) instead of CCS. Algae cultivation is one of the option for biological digestion. Environments including CO_2 concentration are important for algae growth. However, there were few discussions about the methods to recover CO_2 and to separate it before utilisation in the integrated process, and about the quantitative estimations in terms of energy consumption.

In this study, suitable algae growth environments have been reviewed for CO_2 capture in the first step. Then, the authors proposed a suitable CO_2 separation method by algae and evaluated its performance and energy consumption quantitatively by experiments and simulations for the integration of low CO_2 sources, the membrane separation process, and algae cultivations as CO_2 capture and utilisation.

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2. Review of algae cultivation environments for CO_2 utilisation

To select suitable CO_2 concentration methods, proper environment to grow some algae species was studied by literature review.

Firstly, the authors searched the publications, which treated the appropriate pH of medium for algae growth. Komatsu et al. (1994) reported that the alga *E. Prolifera* grew well at pH 6.5 while ventilating pure CO_2. In addition, the alga grew best at pH 8.5 and died at pH 4 or less as Figure 1 shows. Some studies report the best growth at near pH 8 (Takahashi et al., 2012). On the other hand, in the case of an oxyphilic alga such as *Chlamydomonas eustigma* (Guillermo E., 2018), the alkalisation process will not be required. Thus, a simpler process without an acidic gas removal unit is expected in this case.

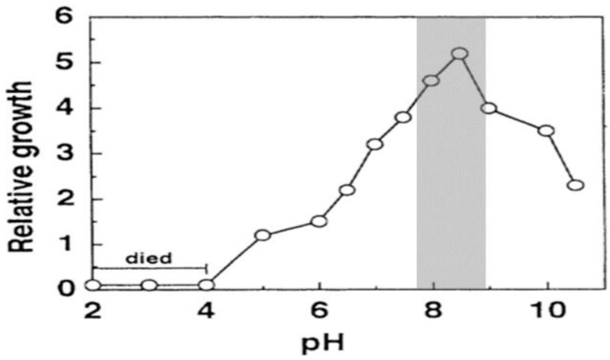


Figure 1: Dependence of cultivating pH for E. Prolifera with ventilating pure CO_2 (Komatsu et al., 1994)

Takahashi et al. (2012) reported that the concentration of nitrogen significantly affects to the cultivation of a blue alga and a diatom (*Microcystis aeruginosa* / *Cyclitella* sp.). In addition, they reported that the algae cell density was increasing with the feeding quantity of nitrate ion, NO_3^-. In this research, the authors concluded that it is essential to feed nitrate ions for some algae cultivation.

Furthermore, there are several publications, which reported the effect of CO_2 concentration to the algae cultivations. Some of them reported experiment with ventilating air containing CO_2 to promote algae growth. These results are summarized in Table 1. In most of them, a low level of CO_2 concentration (2-15 %) was used. Komatsu et al. (1994) reported CO_2 fixation efficiency was only 9 % when they provide pure CO_2 (99.9 %) to the alga shown in Figure 2. It seems that most of the feeding CO_2 did not dissolve in the water and was released into the atmosphere. Therefore, in the case of adding CO_2 directly to cultivate algae, it is not necessary to concentrate CO_2 to high purity.

Table 1: CO_2 concentration in ventilating CO_2-included air to some algae cultivation

Alga species	Conc. of CO_2 in feed gas [%]	Reference
Scenedesmus dimorphus	2 %	Iwai et al., 2017
Botryococcus braunii	5 %	Murakami et al., 1996
Minami-aonori	15 %	Kishimoto et al., 2012
E.Prolifera	99.9 %	Komatsu et al., 1994

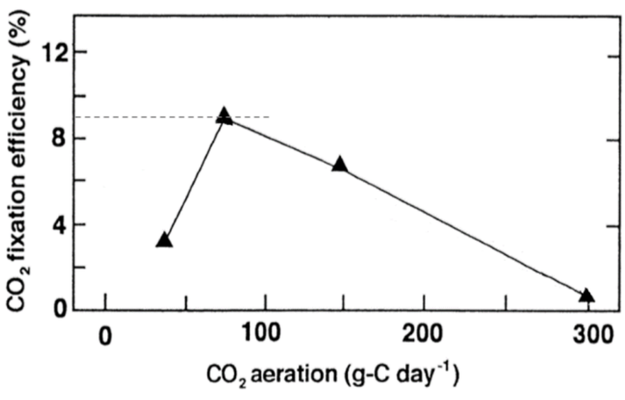


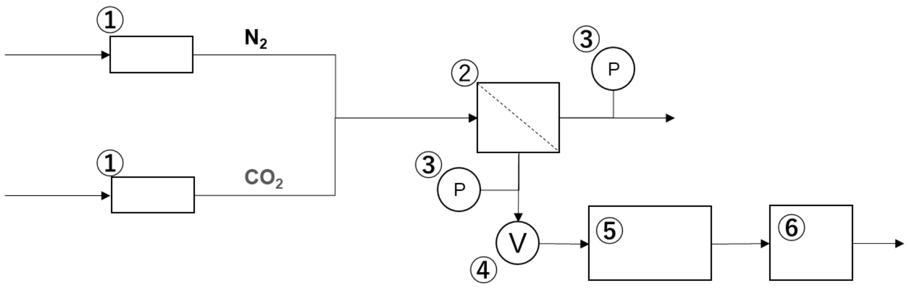
Figure 2: CO_2 fixation efficiency as a function of CO_2 aeration (Carbon-based flow rate) (Komatsu et al., 1994)

3. Membrane separation performance evaluation

A zeolite membrane separation was selected due to the low energy characteristics with algae cultivation environments. The membrane separation performance was experimentally examined, and energy performance was evaluated quantitatively by a commercial process simulator (PRO/II ver.10.2) with the experimental results.

3.1 Separation performance of the zeolite membrane

The schematic image of the experimental setup is shown in Figure 3. Two mass flow controllers (Fujikin Inc. FCS-PM1000A-SP) were set to the system to control the flow rate of each gas, CO_2/N_2. Unit number 2 in Figure 3 is a separation unit. A tubular-type membrane is made of zeolite in this unit. High purity N_2/CO_2 (purity 99.99 %) gases were used.
A needle valve was used to adjust feed pressure. In the steady-state of the gas flow, CO_2 concentration in permeated gas was measured by FI-IR (Thermo Fisher Scientific K.K., Nicolet iS5 iD1 transmission). Flow-meter (Ellutia 7000 series) to check the gas flow rate was set. The membrane performance (permeance, k_m) was calculated from these measured values (flow rate, CO_2 concentration in non-permeated side, and each pressure gauge). Permeated gas concentration and flow rate were calculated from the material balance of each gas. The calculation equation is shown in Eq (1). Experimental conditions are summarized in Table 2.



① Mass flow controller
② Zeolite membrane in the separation unit
③ Pressure gage
④ Needle valve
⑤ FT-IR
⑥ Flow-meter

Figure 3: Experimental setup

Table 2: Experimental condition for the membrane performance estimation

CO_2 concentration in feed gas [%]	Separation temperature	Differential pressure
10		
30	30 °C	0.10 MPa
50		

$$N_m = \frac{F_{perm.,m} \cdot x_{perm.,m}}{A} = k_m(p_{feed,m} - p_{perm.,m}) \quad (1)$$

The results of CO_2 concentration in permeated gas flow was summarized in Table 3. It was impossible to condense to high purity with a single step; however, it is possible to condense CO_2 to 1.1-1.3 times higher concentration. In other words, it was confirmed that it is capable of a little separation of CO_2 by the zeolite membrane unit.

Table 3: CO_2 and N_2 concentration in permeated gas flow

CO_2 Conc. in feeding flow [mol%]	Conc. in permeated flow [mol%]	
	CO_2	N_2
10.0	11.1±0.3	88. 9±0.3
30.0	35.9±0.9	64.1±0.9
50.0	64.8±1.3	35.2±1.3

3.2 Energy performance evaluation

Installing the experimental data in section 3.1 into the simulator, the performance of membrane separation was examined. The simulation conditions were as follows: feeding concentration was 10 $_{mol}$% assumed low CO_2 sources, differential pressure and temperature were the same as the experiment, adiabatic efficiency of the compressor to separate the feeding gaseous stream were assumed to be 80 %. Permeance area was assumed 1000 m^2, the value suit approximately 30-50 m^3 scale plant by calculating from the area density 20-30 m^2/m^3 (Strathman et al.,1985). The feed flow rate was changed between 0.01-10 kmol/s. The simulation result is shown in Figure 4 (a). In this figure, it can be seen that energy consumption for CO_2 separation was less than 1 MJ/kg-CO_2 while feed rate was less than 0.03 kmol/s. Moreover, energy consumption was increasing with increasing feeding flow rate. In addition, the recovery ratio was decreasing with increasing of the feed rate. It can be seen that the more flow rate increases, the more it is disadvantageous from an energy consumption and a recovery ratio point of view. CO_2 concentration in the permeated side as a function of feed flow rate is shown in Figure 4 (b). The concentration was increasing but saturating at nearly 19 % over 0.1 kmol/s feeding. In other words, there is no reason to work under a feed rate that is too high.

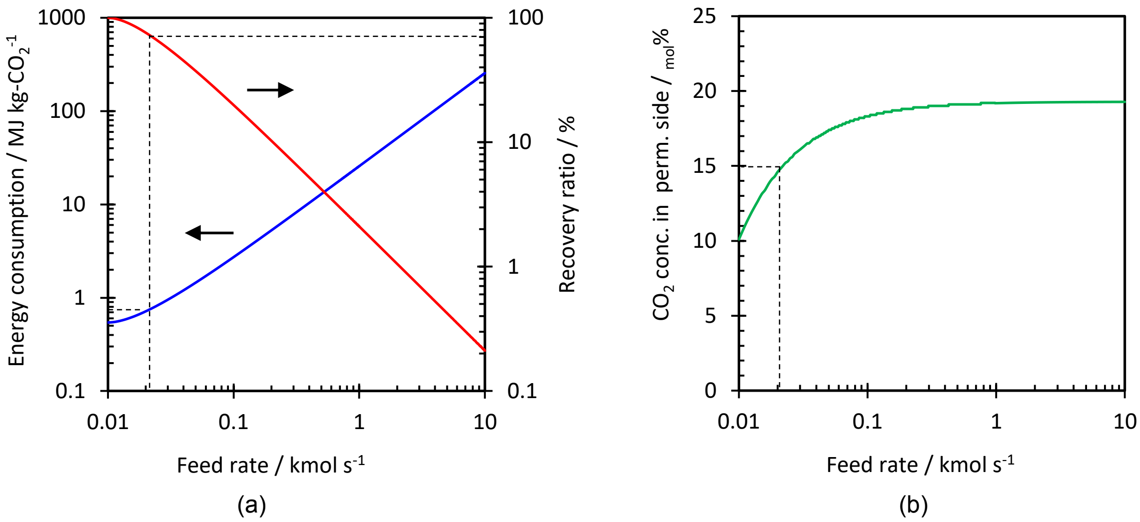


Figure 4: Simulation result (a) energy consumption and recovery ratio (b) CO_2 concentration in permeated gas as a function of feed rate

4. Suggested process description for algae cultivation

Considering these experiment and simulation results of the appropriate algae growth environments investigation and the zeolite membrane performance, an integrated process with the membrane separation was developed. Generally, an exhausted stream from a thermal power generation plant or a garbage incinerating facility includes acidic gases such as NO_x, SO_x, and H_2S. In addition, CO_2 is also known as an acidic gas. Therefore, it is impossible to feed them directly into the medium to decrease its pH. However, some of them are also available for algae growth. Therefore, they should be neutralised by supplying a basic material while they are fed into the medium or be alkalised previously. For the latter method, the reaction equation in the case of feeding a calcium oxide (CaO) as an absorbent is shown in Eq (2). When feeding calcium carbonate ($CaCO_3$) into the medium, carbonate ion (CO_3^{2-}) consumes hydrogen ion (H^+). As a result, the medium pH increase. In the hydrolysis reaction shown in Eq (3), it is expected that alkalisation of the medium and supplying bicarbonate ion (HCO_3^-) and a mineral (Ca^{2+}) occurs simultaneously.

$$CO_2 + CaO \rightarrow CaCO_3 \quad (2)$$

$$CaCO_3 + H^+ \leftrightarrow Ca^{2+} + HCO_3^- \quad (3)$$

However, about NO_x exhausted gas from a thermal power generation plant or a garbage insincerity facility, there are possibilities to utilise for algae growth as a nitrogen source. Nitrate oxide (NO_2) can dissolve into water. The NO_2 decomposes into water and generates nitrate and nitrite ion. These ions are good for the algae.

The zeolite membrane unit could separate CO_2 to a low level of concentration with small energy consumptions. The separation conditions to work the unit should be decided in response to the different purposes in accordance with the utilisation. From investigation results, it was adequate with 15 % CO_2 for algae cultivation. Therefore, the appropriate feed rate was approximately 22 mol/s at this concentration from Figure 4 (b). Then, energy consumption was 0.76 MJ/kg-CO_2, and the recovery ratio was 71 % at the feed rate of 22 mol/s from Figure 4 (a). The result showed energy-saving separation compared with other methods such as 4.0 MJ/kg-CO_2 by a chemical absorption (Goto et al., 2009), 3.8 MJ/kg-CO_2 by a TPSA (Patricia A.P. M. et al., 2017), and a cryogenic method with its phase transition. The process of applying the membrane unit to algae cultivation is promising as one of the CCU technologies. However, water inhibits CO_2 to permeate through the membrane because H_2O molecular polarity is strong compared to CO_2. There is a report to show the performance of each gas with H_2O shown in Table 4 made by the report (Okamoto, 1998). Summarizing the above comprehensively, Figure 5 shows the schematic image of the proposed process for CO_2 capture using algae. The process is firstly removing acidic gases. At this time, NO_x is removed as a nitrogen source for utilising the alga growth. Afterward, H_2O is removed before separating CO_2. Finally, the products are fed to the algae after alkalising the obtained CO_2 and NO_x.

Table 4: Permeance of CO_2/N_2/H_2 gas with/without H_2O molecule

	Permeance, k_m [cm^3(STP) cm^{-2} s^{-1} Hg^{-1}]	
	H_2O	CO_2/N_2/H_2 etc.
Wet (with)	8×10^{-3}	$< 10^{-6}$
Dry (without)	-	$2.5\text{-}3\times10^{-5}$

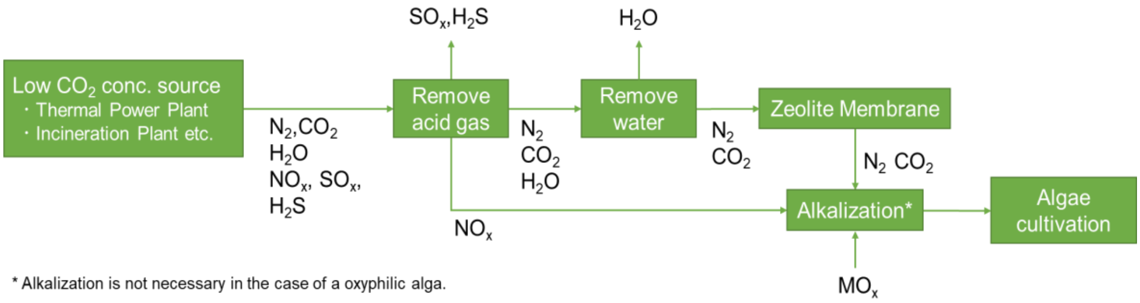


Figure 5: Suggested process considering proper environment for algae and zeolite membrane performance

5. Conclusions

The authors searched the appropriate environment for algae from previous literatures in the first step. The appropriate environments for some algae were as follows; CO_2 concentration was sufficient with 2-15 %. Moreover, the pH was at 8-8.5. However, some algae survive in an acidic environment for their growth. From the results, the authors considered that the membrane separation is suitable for CO_2 separation for feeding algae. Thus, the authors evaluated zeolite membrane separation performance quantitatively by experiments and simulations for their integration and exhaust stream from low CO_2 sources in the next step. The performance of zeolite membrane separation which has been expected as an energy-saving process was evaluated experimentally. It was possible to separate CO_2 to 1.1-1.3 times at 10-50 % CO_2 concentration in feeding streams. By using the experimental result, the energy consumption and the recovery ratio were simulated. As a result, at 22 mol/s feeding stream, the recovery ratio and energy consumption to separate CO_2 were 71 % and 0.76 MJ/kg-CO_2, and this result is realistic and energy-saving compared with other separation methods such as chemical absorption with an amine, physical adsorption, and cryogenic method. Considering the above, the process after recovering low CO_2 from a thermal generation plant or an incineration facility is recovered; separating by membrane unit, applying to algae cultivation is promising in terms of energy consumption. In addition, the overall suggested process description for CO_2 fixation to algae was proposed. The authors recommend applying the membrane separation technology to CCU such as algae cultivation or CO_2 adding agriculture, which requires low CO_2 concentration for the future process design.

Nomenclature

pH – potential of hydrogen
N_m – molar flux of a gas m ($= CO_2/N_2$), mol/(s·m^2)
$F_{perm.,m}$ – molar flowrate in permeated side, mol/s
$x_{perm.,m}$ – molar fraction in the permeated side, -
A – permeation area, m^2

$p_{feed,m}$ – partial pressure in feed side, Pa
$p_{perm.,m}$ – partial pressure in permeated side, Pa

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