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The optimized study of A novel attached membrane photobioreactor on advanced carbon sequestration of microalgae

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ABSTRACT

In this paper, the molecular dynamic study (MDS) and experimental study are conducted to investigate the CO_2 transmission discipline and to improve the carbon sequestration of the microalgae attached membrane photobioreactor (AM-PBR). First, for simulation part, the polyvinylidene difluoride (PVDF) membrane and polytetrafluoroethylene (PTFE) membrane are chosen for MDS. A three-layer simulation model is developed to describe the transmission process of CO_2 from gas-liquid layer to microalgae cell. Then, several parameters including diffusion coefficient, concentration profile and velocity profile of CO_2 is analyzed to compare the different membranes and working conditions. Finally, the experiments are conducted and the AM-PBR is designed to verified the simulation results. The results showed that the CO_2 transmission and diffusion in PVDF membrane is better than PTFE membrane, and the average velocity of CO_2 in PVDF system is 3-9 times higher than the velocity in PTFE system. For different working condition, the CO_2 diffusion in 5% concentration is the highest comparing with other groups in both PVDF and PTFE systems. The experiments proved that the AM-PBR has higher microalgae growth rate than the normal PBRs. This work is a fundamental for the optimization of the PBR and membrane to improve the carbon sequestration rate.

Keywords: microalgae, carbon sequestration, membrane photobioreactor, molecular dynamics

NONMENCLATURE

Abbreviations	
AM-PBR	Attached membrane photobioreactor
DPPC	Dipalmitoylphosphatidylcholine
MDS	Molecular dynamics study
MSD	Mean square displacement
NVE	Microcanonical ensemble
NVT	Canonical ensemble
PBR	Photobioreactor
PVDF	Poly (vinylidene difluoride)
PTFE	Polytetrafluoroethylene
Symbols	
Å	Angstrom
D	Self-diffusion coefficient
N	Number of diffusion particles
r_i	Position of particle
v	Average absolute velocity

1. INTRODUCTION

As promising carbon sequestration and bioenergy production platform, microalgae have received extensive attention in recent years. For microalgae cultivation, different types of photobioreactors are studied for improving the growth rate and carbon sequestration rate of microalgae.[1] However, due to the gas-liquid mass transfer resistance, the carbon sequestration rate of the PBRs is still not up to the expected level. To solve this problem, attached cultivation of microalgae with a membrane is one of the most efficient ways for carbon sequestration and bioresource production.[2]

In the previous study, simulation and optimization of microalgae growth in different kinds of photobioreactors are one of the key issues to improve the growth rate and carbon fixation rate of microalgae[3][4][5]. The photobioreactor for algae cultivation can be divided into open ponds and indoor closed photobioreactor[6]. For open ponds, they are more suitable for large area cultivation. The advantage of this method is its low cost and convenient operation. In this way, direct sunlight is often used to provide the light source needed for microalgae growth. However, the microalgae growth

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rate and carbon sequestration rate of open ponds are low because it is difficult to control the growth environment of microalgae[9]. Therefore, closed photobioreactors are designed for higher growth rates and precise control of the microalgae growth environment. In the previous study, researchers have developed and optimized different kinds of photobioreactor including flat-plate PBR[10], column PBR[11], tubular PBR[12] and membrane PBR[13]. To improve the gas-liquid two-phase distribution and the light-dark cycle characteristics of the photobioreactor, optimized structures such as air-lift spoilers, vortex generators et al. are added on photobioreactors[14]. Although these methods are proposed, due to the gas-liquid mass transfer resistance, the carbon sequestration rate of the PBR is still not up to the expected level. Based on this, the attached membrane photobioreactor is proposed.

For AM-PBR, because of the application of a gas-liquid attached membrane, the CO_2 molecular can be attached directly with the microalgae cell[7]. In this way, the CO_2 utilization rate significantly increases. The main advantages of membrane PBR can be described as: 1) the attachment of CO_2 and microalgae is more close and the carbon sequestration rate can be improved; 2) due to the membrane attachment, the energy consumption of the AM-PBR is relatively low comparing with the flowing PBR; 3) the operational condition of the membrane photobioreactor is easier and don't require additives; 4) because the microalgae grow on the membrane, it is easier to harvest the microalgae by changing the membrane; 5) the membrane photobioreactors are usually assembled in modules which is easy to scale-up and make an applicable carbon sequestration system. Except for these advantages, membrane photobioreactor also has some problems which need to be solved yet. For example, membrane fouling, low membrane lifespan and low selectivity and permeance are the most important problems among them.

Previously, the growth and carbon sequestration performance of the AM-PBR under different working conditions were studied. Rossignol et al. [16]have proved the performance of membrane to enhance the productivity of microalgae valuable production. The research compared two different membrane systems with an external membrane ultrafiltration equipment and an inside tubular agar gel layer. The results showed that both systems are adapted to the continuous culture of microalgae in the photobioreactor. Hou et al. [17] developed a novel sequence batch membrane carbonation PBR for microalgae cultivation. The experiments showed that the optimal pore size of the membrane was 30 nm and the microalgae biomass concentration increased comparing with the normal cultivation methods. Cheng et al. [18]designed a novel AM-PBR for closed space CO_2 elimination. The new PBR successfully increase the CO_2 fixation rate from 80 mg L^{-1} h^{-1} to 260 mg L^{-1} h^{-1} and enhance the retention time of the gas phase from 2 s to 10 s. The efficiency of the AM-PBR is significantly improved. Honda et al.[19] have designed a submerged membrane filtration system in a membrane photobioreactor to improve the carbon capture rate and nutrients removal efficiency. The microalgae are continuously cultivated with simulated treated sewage and 1% CO_2 gas. The optimal hydraulic retention time and solids retention time are determined for the highest carbon sequestration rate and highest nutrients removal efficiency. From the research, it can be concluded that microalgae cultivation in membrane photobioreactor has a higher growth rate and carbon fixation rate, which is a benefit for the utilization of microalgae.

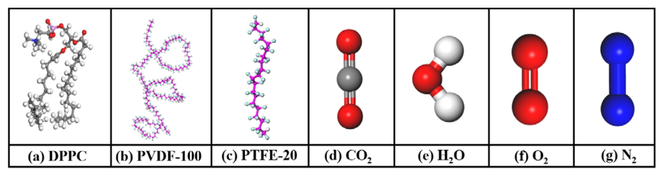


Fig. 1. Model of molecular used in the MDS

However, the AM-PBR also faces the problem of membrane fouling and relatively short lifespan. the fouling problem of the membrane is a crucial bottleneck for the application of membrane photobioreactor[20]. To control the fouling problem of membrane and prolong the lifespan of the membrane, there are several strategies, 1) to modify the characteristics of the membrane and improve the anti-fouling properties, for example, the addition of TiO_2 nanoparticles to the casting solution and use of coarse pore-sized substrates can improve the anti-fouling performance of the membrane[21]; 2) to modify the operation conditions of membrane photobioreactor, such as the gas inlet rate and the CO_2 and nutrients concentration inside the membrane photobioreactor[22],[23]; 3) to modify the structure of photobioreactor, the effective ways such as improving the flow characteristics should be considered for anti-fouling strategies[24].

In the previous studies, researchers have studied the growth and carbon sequestration performance of the AM-PBR, and also employed some modification of membrane or optimization of AM-PBR to solve the fouling problem and improve the growth rate. But the investigation of deeper operation mechanism of AM-PBR is still rare. Manrique et al.[25] have developed the molecular dynamics simulation model for researching the effects of salinity on the CO_2 permeation across lipid bilayer for microalgae bio fixation. The results showed that it is possible to manipulate the MD simulation in membrane PBR. Fuoco et al. [26] have conducted the experiment and simulation work to compare the gas permeation of the highly fluorinated polymer of intrinsic microporosity PIM-2. In conclusion, heat and mass transfer in the attached membrane is one of the complex issues. For these novel and optimized membranes, molecular-level simulation is required for revealing the process. So, revealing the law of the diffusion and mass transfer of gas and nutrients in the membrane are the key issues.

In this paper, the diffusion and transmission performance of CO_2 is analyzed through MDS and experimental methods. First, a three-layer simulation model is built to determine the diffusion coefficient and distribution of CO_2 in different working condition and membranes. Then, the diffusion condition, velocity profile and CO_2 profile concentration distribution inside the system is analyzed. Finally, the experiments are conducted by developing of an attached membrane photobioreactor. The microalgae growth rate and carbon fixation rate of AM-PBR is calculated.

2. MATERAIAL AND METHODS

2.1 Simulation model

For the modeling of the PVDF and PTFE part, PVDF and PTFE polymer and water molecular are involved in the layer. First, the single PVDF and PTFE molecular is developed and according to the literature, when the degree of polymerization is 100 (as shown in Fig.1(b)), the physical properties of PVDF polymer are close to the actual condition. For PTFE molecular, when the degree of polymerization is 20(as shown in Fig.1(c)), the physical properties are consistent with the actual situation. The size of the amorphous cell is 50 Å×50 Å×110 Å. Then, in each PVDF amorphous cell, 14 PVDF-100 molecular and 800 water molecular are involved and the density of the membrane is 1.65 g·cm^{-3}. In each PTFE amorphous cell, the density of the membrane is 2.2 g·cm^{-3}. The membrane amorphous cell is first simulated in the NVT (N: constant particle number, V: constant volume, T: constant temperature) ensemble for 600 ps annealed and 500 ps dynamic calculation. The annealing procedure is to relax the structure to equilibrate the box.

For modeling the microalgae cell membrane, DPPC lipid bilayers and water molecular are involved in the DPPC layers. The DPPC molecular is shown in Fig.1(a) and the structure built in MS software is shown in Fig.1(a). The amorphous cell is composed of 48 DPPC molecular and 1720 water molecular and the density of the membrane is similar to the water density. The size of the amorphous cell is 50 Å×50 Å×55 Å. The same procedure is conducted and the annealing process is calculated.

For the gas layer, the CO_2 molecular, O_2 molecular, N_2 molecular and the water molecular are involved. The specific numbers of molecular in the gas layer are shown in Table 1 and the structures of the gas molecular are shown in Fig.1(d)-(g). The size of the gas layer is 50 Å×50 Å×55 Å and the density of the amorphous cell equals water density. The same simulation process is conducted for the gas layer and the energy of the system reaches the lowest after the annealing process.

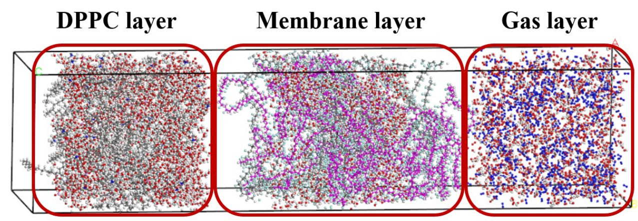


Fig. 2. Three layers model of MDS simulation

Table 1. Different groups of gas layer parameters

No.	Numbers of CO_2	Numbers of N_2	Numbers of O_2	Numbers of H_2O
Group No.1	50	700	300	1000
Group No.2	100	700	300	1000
Group No.3	150	700	300	1000
Group No.4	200	700	300	1000

The constructed three-layer model is shown in Fig.3. The size of the three-layer model is 50 Å×50 Å×220 Å. Four different controlled groups are proposed and calculated separately. The simulation procedure of the layers is as follows: 1) Geometry optimization of the layers with smart algorithm and maximum 500 iterations; 2) Anneal process with 3 annealing cycles. The initial temperature is 300 K and the mid-cycle temperature is 800 K. The annealing process is using NVT ensemble with 500 heating ramps per cycle; 3) Dynamics calculation at 298 K. The process is using NVE (N: constant particle number, V: constant volume, E: constant energy) ensemble for 500 ps simulation. The whole simulation processes use COMPASS II forcefield and atom-based electrostatic and van der Waals summation method.

The molecular dynamics calculation of the layers is conducted through Forcite Anneal and Dynamics. The ensemble is chosen as NVT at 298 K. For anneal process, 3 annealing cycles from 300 K to 800 K are employed with 300000 total steps. The all the MD simulation production employed a total simulation time of 500 ps and the time step is set at 1 fs.

2.2 Experiments methodology

The experimental study is employed in this paper to build the AM-PBR system. The PBR consists of three part, the gas-liquid chamber, the PVDF/PTFE membrane and the microalgae chamber. The gas-liquid chamber and the microalgae chamber is made by plexiglass, the size of each chamber is 200 mm×200 mm×50 mm. The two chambers are connected by screws with membrane between two chambers. The membrane is clamped by the plexiglass to separate the PBR (as shown in Fig. 3).

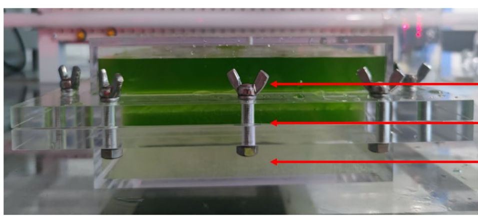


Fig. 3. Physical map of AM-PBR

For the membrane part, a commercial PVDF membrane (MF022) purchased from RisingSun Membrane Technology (Beijing) is first employed in the experiment to test the growth and carbon sequestration performance of the PBR. The pore size of the membrane is 0.22 µm and the size of the membrane is cut to 20 cm × 20 cm.

The PBR system is operated in a self-built artificial greenhouse to maintain the suitable microalgae cultivation temperature. The mixture of CO_2 and air passes through a mass flow meter to supply the appropriate concentration and gas inlet velocity of CO_2 for the growth of microalgae. The microalgae used in the experiments is *Chlorella pyrenoidosa*, purchased from Shanghai Guangyu Biotechnical Co. Ltd. In the gas-liquid chamber, BG11 culture medium is chosen as the growth medium, the specific formula of the BG11 is as follows: 1.5 g·L^{-1} $NaNO_3$, 0.04 g·L^{-1} K_2HPO_4, 0.075 g·L^{-1} $MgSO_4·7H_2O$, 0.036 g·L^{-1} $CaCl_2·2H_2O$, 0.006 g·L^{-1} Citric acid, 0.006 g·L^{-1} Ammonium ferric citrate, 0.001 g·L^{-1} $EDTANa_2$, 0.02 g·L^{-1} Na_2CO_3 and 1 ml A5 trace elements (0.222 g·L^{-1} $ZnSO_4·7H_2O$, 0.079 g·L^{-1} $CuSO_4·5H_2O$, 0.015 g·L^{-1} MoO_3, 0.036 g·L^{-1} $CaCl_2·2H_2O$, 2.86 g·L^{-1} H_3BO_3, 0.006 g·L^{-1} $MnCl_2·4H_2O$)

3. THEORY/CALCULATION

3.1 MDS calculation

The previous three-layer model is prepared for MDS simulation. The simulation procedure of the layers is as follows: 1) Geometry optimization of the layers with smart algorithm and maximum 500 iterations; 2) Anneal process with 3 annealing cycles. The initial temperature is 300 K and the mid-cycle temperature is 800 K. The annealing process is using NVT ensemble with 500 heating ramps per cycle; 3) Dynamics calculation at 298 K. The process is using NVE ensemble for 500 ps simulation. The whole simulation processes use COMPASS II forcefield and atom-based electrostatic and van der Waals summation method.

Table 2. Diffusion rate of PVDF and PTFE systems

No.	Group 1	Group 2	Group 3	Group 4
D_{PVDF} / cm^2·s^{-1}	3.12×10^{-5}	2.40×10^{-5}	2.88×10^{-5}	2.50×10^{-5}
D_{PTFE} / cm^2·s^{-1}	1.53×10^{-5}	1.31×10^{-5}	1.52×10^{-5}	1.15×10^{-5}

3.2 Calculation of diffusion coefficient

The diffusion process of the CO_2 inside the PVDF membrane and the microalgae cell membrane is complex and the diffusion coefficient describes the rate and this process. In the MD simulation, the molecular moves from the initial position and diffuses in the membrane. The mean value of square displacement of a particle is defined as the mean square displacement (*MSD*). The self-diffusion coefficient of the molecular can be calculated through the Einstein law of diffusion, which is shown in eq. (1),

$$D = \frac{1}{6N} \lim_{t\to\infty} \frac{\mathrm{d}}{\mathrm{d}t} \sum_{i=1}^{N} \left[r_i(t) - r_i(0) \right]^2 \quad (1)$$

where D is the self-diffusion coefficient, N is the number of diffusion particles. $r_i(t)$ means the position of particle i at t moment. The output trajectory document records the diffusion trajectory of particles and *MSD* change with time. The average number of particles is calculated and based on this, the self-diffusion coefficient D can be directly calculated according to the slope of the *MSD* curve.

3.3 Calculation of profile concentration

The distribution of CO_2 inside the membrane and the microalgae is the key issue to evaluate the carbon sequestration and the CO_2 absorption performance of the PVDF membrane. In the MD simulation, the profile concentration shows the relative concentration of a molecular along with different directions. In order to compare the performance of different controlled groups, the profile concentration is analyzed. The concentration profile calculates the concentration of particles in a given layer. In the calculation process, the profile concentration of the *z*-axis is analyzed to determine the CO_2 distribution in different layers.

3.4 Calculation of velocity profile

The velocity of CO_2 inside the membrane is an important factor to measure the diffusion rate. The velocity profile contains the average velocity component of the particles in a given direction. In the MD simulation, the trajectory file is generated and the velocity profile of CO_2 molecular is analyzed. In the calculation process, the velocity profile of the *z*-axis is analyzed to determine the CO_2 distribution in different layers.

Table 3. Average relative concentration of CO_2 in each layer

No.	Gas layer	PVDF layer	DPPC layer
PVDF Group 1	0.845	0.706	1.774
PVDF Group 2	0.849	0.912	1.346
PVDF Group 3	1.154	0.770	1.280
PVDF Group 4	1.146	0.798	1.270

3.5 Calculation of carbon sequestration rate

The volumetric growth rate of microalgae is determined by the biomass concentration change in a certain time interval, as shown in eq. (2):

$$P = \frac{c_{t+\Delta t} - c_t}{\Delta t} \quad (2)$$

where $c_{t+\Delta t}$ and c_t represents the microalgae biomass concentration at times *t* and *t+Δt*, and the unit of them is $g \cdot L^{-1}$. *Δt* is the time period of the specific growth rate, and the unit is seconds.

The carbon fixation rate of microalgae can be evaluated from the carbon content inside the algae cell and the volumetric growth rate of microalgae, as shown in Eq. (14):

$$R_C = PC_C \left(\frac{M_{CO_2}}{M_C} \right) \quad (3)$$

where C_C is the carbon content inside the microorganism, and M_{CO_2} and M_C represents the relative molecular weight of CO_2 and C, which are 44 and 12. The value of C_C is obtained from the literature and determined to be 51% [27].

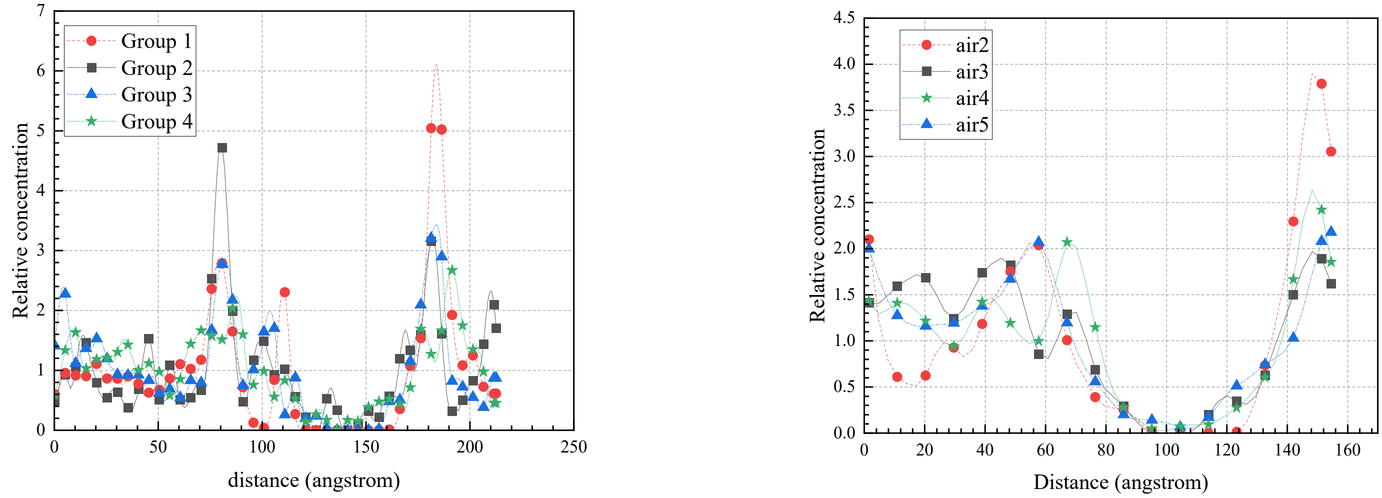
4. RESULTS AND DISCUSSION

4.1 Diffusion coefficient analyzation

The simulation is completed according to the aforementioned methods and simulation models. The diffusion coefficients of different controlled groups shown in Table 1 are analyzed. The results are represented in Table 2. It can be figured that the *D* of PVDF membranes are generally higher than the PTFE membranes. For the four different condition, the D_{PVDF} in Group No.1 is 30% higher than that in Group No.2. This indicates that even though the concentration inside the cell membrane is higher, the diffusion and CO_2 transmission inside the PVDF membrane is worse as the CO_2 concentration increase. The reason is that at higher CO_2 concentrations, the pass of CO_2 is difficult due to the interaction force of the molecules. Since that the attached microalgae on the PVDF membrane needs a carbon source for photosynthesis, Group No.1 is the most suitable condition for microalgae growth and carbon sequestration.

4.2 CO_2 concentration distribution

Fig. 5 shows the profile concentration along the z-axis of CO_2 inside the layers at different distances after 500 ps concentration. It can be figured that the average concentration of CO_2 distributed in PVDF group is higher than PTFE group. The average relative concentration after 500 ps in each layer is shown in Table 3. In Fig. 4(a), the four lines represent the relative concentration in different groups of PVDF membrane. The results showed that increasing the concentration of CO_2 in the gas layer is not beneficial for the carbon sequestration in the DPPC layer. The average relative concentration inside the microalgae cell membrane is lower at a higher concentration. According to the previous research, the microalgae growth rate changes with CO_2 concentration, and the optimal concentration for carbon sequestration is 4%. When the concentration increases in the range above 4%, the microalgae growth rate decreases. On the other hand, the higher concentration of the gas layer leads to better diffusion in the PVDF layer. It can be figured that although the average relative concentration of CO_2 inside the PVDF membrane is barely any different, the variance of the relative concentration is extremely different. The variance of the first group is 0.777 and the variance of the fourth group is 0.374. It indicates that the



(a) PVDF membrane (b) PTFE membrane

Fig. 4. Relative concentration of CO_2 in the layers

uniformity of the CO_2 gas distribution in the PVDF membrane is better in Group No.4.

4.3 Velocity profile analyzation

The average velocity profile of CO_2 in multi-layer system in 500 ps simulation is analyzed. In Fig. 5, the results of the four different controlled groups are shown. The average absolute velocity of the four groups is shown in Table 4. It can be figured that the velocity in PVDF membrane is much higher than the velocity in PTFE membrane. The average velocity of Group 1 is the highest in the four groups which are 91.7% higher than the lowest Group 4.

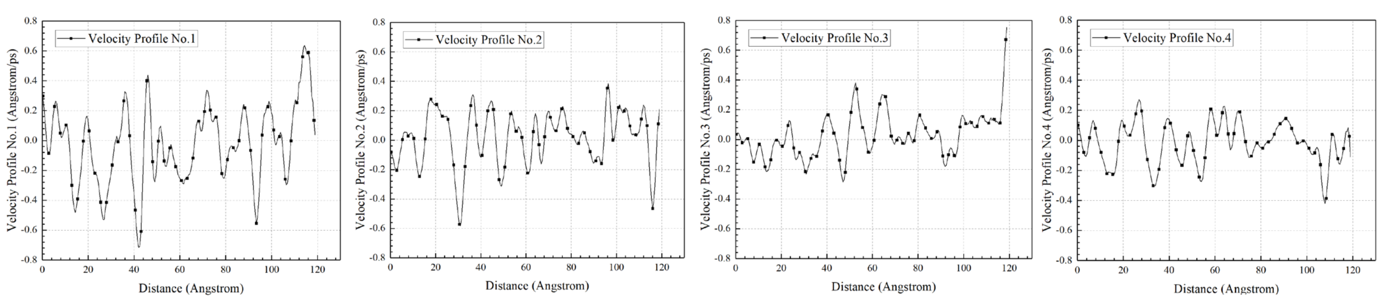
Table 4. Average absolute velocity of CO_2

Group No.	Group 1	Group 2	Group 3	Group 4
V_{PVDF} / Å²·ps⁻¹	0.207	0.148	0.111	0.108
V_{PTFE} / Å²·ps⁻¹	0.071	0.042	0.016	0.015

From the analyzation above, it can be concluded that the PVDF membrane has better performance in concentrating CO_2 molecular and evenly distribution of CO_2 gases in membrane layer and microalgae layer. Therefore, in the experimental study, PVDF membrane is chosen for further research.

4.4 Experimental research analyzation

The continuous cultivation of microalgae for 3 days is employed in the PBR. The microalgae concentration in the PBR is tested every 8 hours and after the cultivation, the membrane is drying to determine the dry weight and the microalgae attached on the membrane. The carbon sequestration rate of AM-PBR is calculated by eq.(3), which is 4.48 mg·h^{-1}. The biomass concentration changes in the microalgae chamber is shown in Fig. 6. It proves that the membrane system can be used for microalgae cultivation. In this system, because of the gas permeation rate of commercial PVDF membrane is low, although the growth rate and carbon sequestration rate of the AM-PBR are higher than normal PBR, but it is not optimistic for AM-PBR. Therefore, to increase the performance of AM-PBR, self-made modified PVDF membrane is needed and due to the time limitation, the experiments are still in process. In the following study, the porogen and



(a) Group 1 (b) Group 2 (c) Group 3 (d) Group 4

Fig. 5. Average velocity profile along the z-axis of CO_2 molecules in 500 ps

hydrophilic modification of the membrane will be applied to improve the gas permeation rate and the attach of microalgae on the membrane.

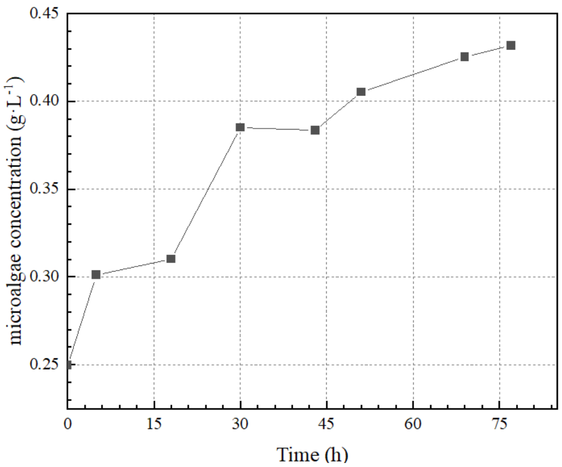


Fig. 6. Biomass concentration changes in AM-PBR

5. CONCLUSIONS

In this paper, the diffusion and distribution performance of CO_2 in PVDF and PTFE based AM-PBR system is determined and the PBR is employed in the experimental study. A multilayer system with different groups of CO_2 concentration is proposed. The diffusion coefficient, concentration profile and velocity profile of CO_2 are calculated. The experimental study with a commercial PVDF membrane is conducted and the carbon sequestration ability of the PBR is verified preliminarily. The main conclusions in the paper are as follows:

(1) According the simulation results, the CO_2 transmission and diffusion performance of PVDF membrane is better than PTFE membrane. For PVDF membrane, after annealing and 500 ps simulation, the relative concentration of CO_2 in membrane layer and microalgae layer is higher and the diffusion coefficient of CO_2 is higher.

(2) For different working condition, in four controlled groups, the diffusion coefficient is higher in Group 1, which has a lower CO_2 concentration. And with the increase of the CO_2 concentration, the diffusion coefficients decrease. Also, the average absolute velocity of CO_2 is higher at lower CO_2 concentration.

(3) According to the experiments, the growth rate of the microalgae is improved comparing with the normal PBR without membrane. The membrane still needs improvement and modification.

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