Article

Study of Aeration and CO_2 Absorption Using Filtration Membranes in Terms of Physical Properties and Mass Transfer Parameters

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Abstract. Hollow fiber membrane contactor is nowadays one of the alternative absorption processes for the conventional bubble column. In this work, the performances of microporous hollow fiber membranes for aeration and CO_2 absorption were investigated and compared with a conventional bubble column. The membranes used in this work were adapted from filtration membranes which are significantly cheaper than conventional absorption membranes. The effects of operating variables such as average pore sizes, gas flow rates, liquid flow rates, number of hollow fiber membrane, and concentrations of chemical solution on the gas-liquid absorption rate were determined. For oxygen-water absorption, the overall mass transfer coefficient (k_La), which corresponding to the absorption rate, increased with the increase of membrane porous diameter. The k_La was also enhanced with the increase of the liquid flow rate and the number of membranes while the gas flow rate was rarely influent. For carbon dioxide absorption, the increase in liquid flow rate and the carbon dioxide concentration resulted in higher mass transfer rate. Moreover, adding sodium carbonate in absorbent improved the k_La value up to 2.2 folds, comparing with physical absorption. The comparison between the membrane contractor and a bubble column indicated that the utilization of filtration membranes had more efficiency comparing to bubble column due to its high surface area and adaptability when operating with the same size especially when using lower gas velocity rate.

Keywords: Gas absorption, hollow fiber membrane contactors, aeration, CO_2 absorption, filtration membranes.

1. Introduction

Carbon dioxide (CO_2) is, nowadays, one of the concerning substances for global warming. The concentration of CO_2 in atmospheric has dramatically increased to approximately 450 ppm by 2030 and between 750 ppm and 1,300 ppm by 2100 [1]; Increasing of CO_2 in atmospheric causes global warming, climate change, and subsequently affects creatures in earth. The major cause of the increase of CO_2 in atmosphere is the combustion of organic substances that used for energy generation, electricity, and industry processes. In order to prevent the exposing of CO_2, the conventional equipment for CO_2 absorption were pioneered including bubble column, spray column, packed column and tray column. All of the operations provide contacting areas between gas and liquid to transfer CO_2 from gas phase to liquid phase in order to reduce the amounts of CO_2 exposing to atmosphere. From all equipment, bubble column is widely used because of its ease in operation procedure, absorption capacity as well as its low operating cost. However, industrial-scale bubble columns are confronted a variety of operational problems including flooding, entrainment and foaming [2]. Hence, in this recent years, the gas absorption using hollow fiber membrane contactor has been investigated to be an alternative of the bubble column; not only it can provide greater gas-liquid transfer area comparing to the conventional equipment [3] but also its operational flexibility, economic operating cost, linear scale-up, and simple prediction [4].

The success of using hollow fiber membranes for CO_2 removal can be found in many researches. For CO_2-water absorption, various removal efficiencies were reached, depending on their operating conditions and material of membranes [5-9]. The maximum CO_2 removal of 76% by the absorption in water was achieved in a polypropylene hollow fiber membrane [5], while only 20% of CO_2 removal efficiency was reached with a polyvinylidenefluoride membrane [6]. The improvement of CO_2 removal could be done by introducing the chemical reaction that could enhance the treatment efficiency up to 87% when the monoethanolamine was used with a polypropylene membrane [7].

The membranes used in the membrane contactor can be hydrophobic or hydrophilic and its pores can be either gas filled or liquid filled [5, 10]. Hydrophobic surface membranes with high porosity and chemical resistance was claimed to be the most effective one [11]. It was also found that the throughput of liquid in the tube side offers a significant improvement in performance, comparing when operating in the shell side of the module [3]. Normally, specific absorption membranes are made of polypropylene (PP) and Polytetrafluoroethylene (PTFE). Regarding the hollow membrane required very specific characteristics (i.e. chemical resistance, high porosity etc.), the cost of production for these membranes are very high [11]. In order to overcome the problem, in this work, the membrane using for filtration process was adapted to use in absorption system since filtration membranes also have high porosity, chemical resistance with significantly lower cost.

Therefore, this research purpose was to investigate the performances of hollow fiber filtration membranes and compared their performance with conventional bubble column in term of the overall volumetric mass transfer coefficient (k_La). The specific design criteria and the suitable correlation for predicting the performance of hollow fiber membrane contactors for gas absorption of O_2 and CO_2 were identified.

2. Methodology

2.1. Membrane Characterization

In this research, a polyvinylidene fluoride microfiltration membrane (MF), represented as type A, and two Ultrafiltration membranes (UF), represented as type B and C, were studied. Their physical characteristics were identified with an environmental scanning electron microscope (ESEM). The Wilhelmy method was applied to measure the contact angle of the membranes surfaces [11].
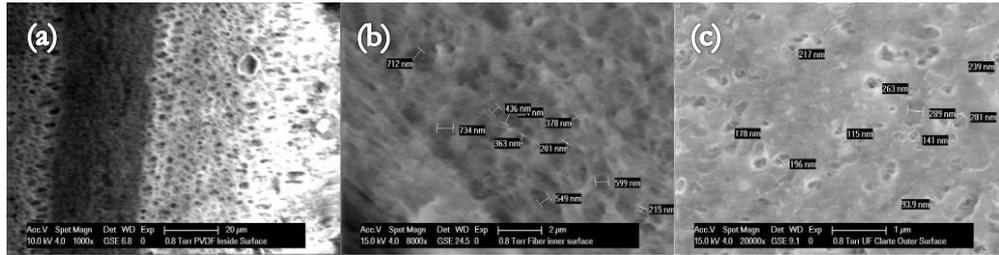
Fig. 1.   Membrane surface morphology (a) Membrane A (b) Membrane B (c) Membrane C.

The structures and surfaces of membrane A, B and C identified with ESEM are illustrated as in Fig. 1. The surface of membrane A was smooth as can be seen in Fig. 1(a). The porous diameter of membrane A was 0.160 μm; the contact angle of membrane A was 61.1 which indicated that the surface of membrane A was hydrophilic. The membrane B and C consisted of fibers (Type B) and small porous (Type C), as shown in Fig. 1(b) and 1(c), had their porous diameter to 0.024 and 0.109 μm, respectively. The property of membrane B and C were hydrophobic and hydrophilic corresponding to their contact angle of 136.0 and 0.0, respectively. The summary of the characteristics of hollow fiber membranes are provided in Table 1.

Table 1.   Characteristics of hollow fiber membrane used in this research.

Type	Porous diameter (μm)	DCA (°)	Hydro-property
A	0.160	61.1	Hydrophilic
B	0.024	136.0	Hydrophobic
C	0.109	0	Hydrophilic

2.2. Experimental Set-Up

In this work, both aeration and CO_2 absorption were studied. The equipment used in the aeration process were set up as illustrated in Fig. 2. The hollow fiber membranes were placed inside a 1-cm inside-diameter and 20-cm effective length glass module. Air was fed to the shell side of the glass module with air pump and regulated its flow rate with a flow meter. The air flew pass the outer surface of hollow fiber membrane before exposed to atmosphere. The inside of hollow fiber membranes were connected to liquid pump in order to circulate liquid inside membrane and liquid circulation system. The counter current regime was selected as it provided a great driving force for mass transfer [3]. Oxygen-free water was prepared by the chemical reaction of sodium sulphite (Na_2SO_3) with dissolved oxygen in water [12]. Within the liquid flow circulation line, a dissolved oxygen sensor, Horiba OM-51, was used to measure oxygen concentration in liquid phase as function of time.

For carbon dioxide experiments, experimental set-up was almost the same as aeration system exclude gas feeding system and the measurement methodology. Noted that for CO_2 absorption system, the gas phase was not continuously flow but accumulated within the shell side of module due to the limitation of measurement. The initial CO_2 concentration at 9000 ppm was prepared by mixing pure CO_2 with fresh air before initiated to the shell side of membrane. The Testo CO_2-535 was used as the CO_2 concentration sensor inside the reactor in order to determine mass transfer parameters and its efficiency. The deionized water with sodium carbonate (Na_2CO_3) solution at 0, 1, 5, and 10 percent by weight was used as liquid phase for absorption of CO_2.
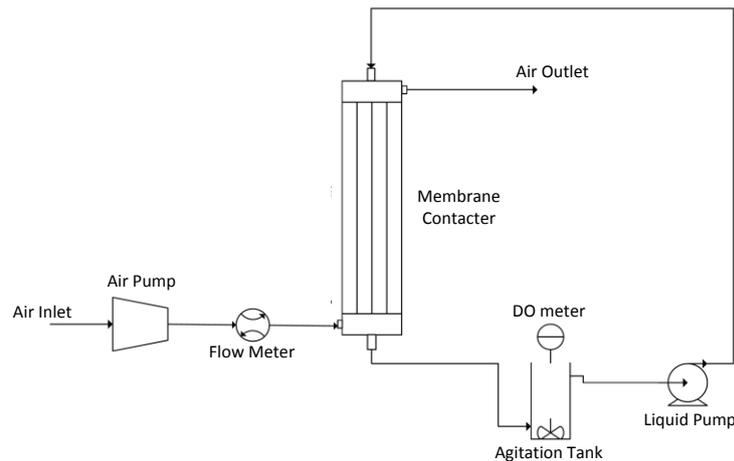
Fig. 2.   Experimental set-up for aeration system.

2.3. Analytical Method

In all experiments, the concentration data was collected every 10 seconds until 30 minutes to ensure that the steady state had been reached. All the experiments were conducted at temperature of 25 °C.

2.3.1. Determination of overall mass transfer coefficient (k_La) in aeration system

The mass transfer coefficient (k_La) for aeration process (O_2 absorption in water) were determined by applying the American Society of Civil Engineers (ASCE) method [13] which derived from mass balance equation as shown in Eq.(1);

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

where its integration form is expressed as in Eq. (2)

$$\ln(C^* - C_L) = \ln(C^*) - k_L a \cdot t \tag{2}$$

From Eq. (2), k_La value was determined from the slope of $\ln(C^*_l - C_l)$ and t.

2.3.2. Determination of overall mass transfer coefficient (k_La) in CO_2 absorption system

The k_La determination method for CO_2 absorption system was different from aeration system because the gas phase was not flow continuously. The method was initially derived with the mass balance equation of CO_2 transferred between gas and liquid phase. At any transfer concentration, the change of dissolved CO_2 in liquid phase must be equal to CO_2 transferred from gas phase. Hence, the CO_2 balance equation is given by the following Eq. (3) and can be written as in Eq. (4) where V refers to volume of fluid in system, C refers to CO_2 concentration, subscripts of g and L refer to gas and liquid phase while i and t are corresponding to initial and at considered time respectively.

$$\Delta C_g V_g = \Delta C_L V_L \tag{3}$$

$$(C_{g,in} - C_{g,t})V_g = (C_{L,t} - C_{L,i})V_L \tag{4}$$

As the initial CO_2 concentration in DI-water was zero, concentration of gas as function of liquid concentration can be expressed as in Eq. (5);

$$C_{g,t} = C_{g,in} - \frac{C_{L,t}V_L}{V_G} \tag{5}$$

Eq. (1) combines with Henry's Law can be written as;

$$\frac{dC}{dt} = k_L a(C^* - C_{L,t}) = k_L a(k_H C_{g,t} - C_{L,t}) \tag{6}$$

Combine Eq. (5) with Eq. (6) yields;

$$\frac{dC}{dt} = k_L a \left[k_H C_{g,in} - C_{L,t} \left(\frac{k_H V_L}{V_G} + 1 \right) \right] \tag{7}$$

Define $\theta = \frac{k_H V_L}{V_G} + 1$, therefore, Eq. (7) becomes;

$$\frac{1}{k_H C_{g,in} - C_{L,t}\theta} dC_L = k_L a \cdot dt \tag{8}$$

$$\ln \frac{k_H C_{g,in} - C_{L,t}\theta}{k_H C_{g,in} - C_{L,i}\theta} = -k_L a \cdot \theta \cdot t \tag{9}$$

From Eq. (9), the $k_L a$ for CO_2 absorption can be obtained from slope of $\ln \frac{k_H C_{g,in} - C_{l,t}\theta}{k_H C_{g,in} - C_{l,i}\theta}$ and θt.

2.3.3. CO_2 Removal efficiency

CO_2 removal efficiency is considered from difference between CO_2 concentration at initial condition and at considered time with initial concentration as divisor as shown in Eq. (10)

$$CO_2 \text{ Removal Efficiency} = \frac{CO_{2,initial} - CO_{2,at\ time\ t}}{CO_{2,initial}} \times 100 \% \tag{10}$$

3. Results and Discussions

3.1. Membrane Selection

The oxygen-water absorption experiment was conducted at gas and liquid flow rate of 10 LPM and 0.1 LPM respectively. The results as shown in Fig. 3 indicated that the $k_L a$ of membrane A were higher than B and C due to its larger porous size. However, membrane B that had smaller porous diameter yielded higher $k_L a$ than membrane C; it was possibly happened because the surface of membrane B was hydrophobic which normally dry during the operation between air and water. This dry operation was confirmed to achieve higher rate comparing with the wet operation of hydrophilic surface [14]. According to its highest mass transfer coefficient, membrane A was selected to study the effect of operating conditions on its mass transfer parameters and efficiency.
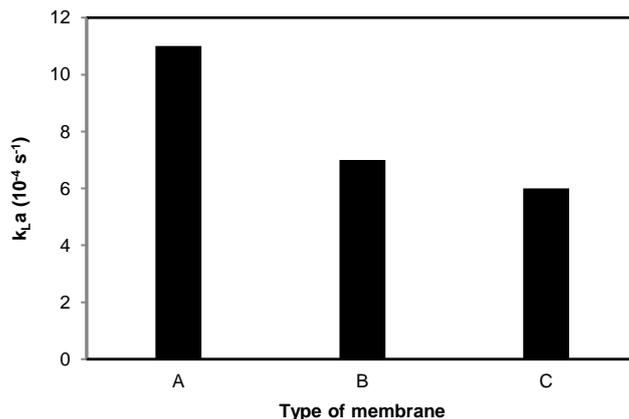
Fig. 3. Comparison of the k_La in aeration system with different types of membrane. (Q_g = 10 LPM, Q_L = 0.1 LPM, Number of membrane module = 5).

3.2. Effect of Operating Variables and Membrane Configuration on the Overall Mass Transfer Coefficient (k_La)

In this part, the effect of operating variables consisted of gas flow rate, liquid flow rate as well as number of membrane on overall mass transfer coefficient (k_La) of membrane A were investigated.

3.2.1. Effect of gas flow rate

Figure 4 illustrates the effect of gas flow rate on the k_La at constant liquid flow rate of 0.1 l/min. In the figure, the k_La value slightly rose as the gas flow rate included which implied that the gas flow rate had slightly effect on the k_La. Due to this fact, it could be interpreted that the mass transfer rate for the hollow fiber membrane was limiting at liquid interphase [6, 15].
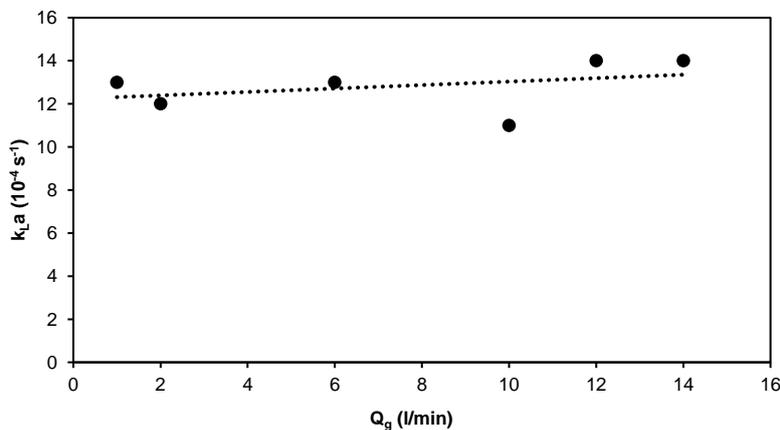

Fig. 4. Effect of the gas flow rate on the overall mass transfer coefficient (k_La) in aeration system (Q_L = 0.1 LPM, Number of membrane module = 5).

Similar results were reported by Wang et al. [15], the gas flow rate was found to have no impact on the overall mass transfer coefficient. Not only for the aeration process, it was also confirmed by Atchariyawut et al [6] that the mass transfer resistance of CO_2–water absorption was limiting in the liquid phase, which normally found in the absorption of low-soluble substances.

3.2.2. Effect of liquid flow rate

Figure 5 shows the effect of liquid flow rate on the k_La at gas flow rate of 2 l/min. As seen in the figure, the increasing in liquid flow rate greatly enhanced k_La value especially at low liquid flow rate. The increment of liquid flow rate caused higher liquid velocity in the hollow membrane, and reduced the mass transfer resistance in liquid phase. The observation is supported by Graetz-Lévêque equation (Eq. 11) which normally used to predict the tube side mass transfer coefficient [16, 17].

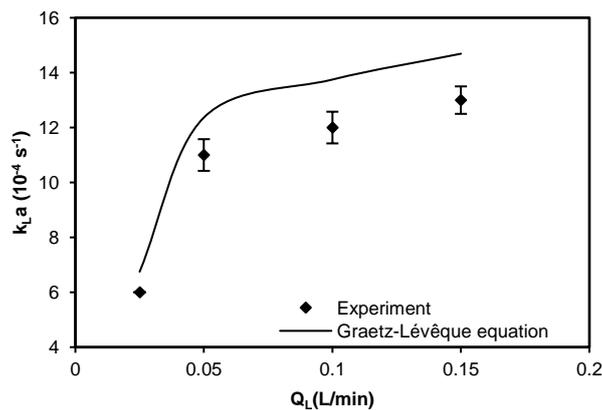$$Sh = \frac{k_L d_i}{D_i} = 1.62 \sqrt[3]{\frac{d_i^2 V_L}{L \cdot D_i}} \tag{11}$$

Fig. 5. Effect of the liquid flow rate on the overall mass transfer coefficient (k_La) in aeration system (Q_g = 2 l/min, Number of membrane module = 5).

The similar behavior was also reported by Wang et al. [15]. It was claimed that the increase of the liquid velocity could accelerate the supply of the active absorbent, increase mixing, and mitigate the depletion effectively. As the result, the gas absorption flux increased with the increase of the liquid velocity. Fortescue et.al [18] and Atchariyawut et al [6] also mentioned this similar effect in term of Reynolds number.

3.2.3. Effect of the amount of hollow fiber membrane

The effect of the amount of hollow fiber membrane is presented in Fig. 6. The result obviously indicated that the k_La roughly increased equal to the increase of the total amounts of membrane used. As can be seen in the figure, applying 5 units approximately provided the k_La at 11 x 10^{-4} s^{-1} while applying 10 units yield the k_La at around 21 x 10^{-4} s^{-1} which was approximately two times higher. It could be clearly explained that, since the overall mass transfer coefficient (k_La) consists with two terms in multiply i.e. liquid side mass transfer coefficient (k_L) and specific interfacial area (a), the increment of unit numbers of membrane significantly increased the interfacial area (a) equals to the times of unit increase and, consequently, rose the k_La [19].

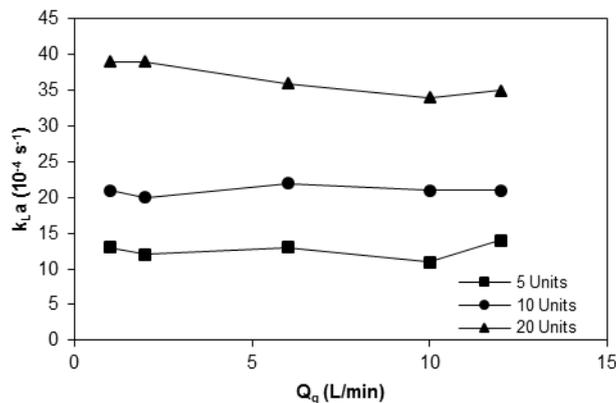Fig. 6.   Effect of number of hollow fiber membrane on the overall mass transfer coefficient (k_La) in aeration system ($Q_L = 0.1$ l/min).

3.3. Comparison the k_La between Hollow Fiber Membrane Contactor and Bubble Column Reactor

The comparison of the bubble column results of Sastaravet et al. [20] with the k_La from membrane type A is depicted in Fig. 7. According to their bubble column, three types of single orifices including flexible (Fo1), rigid (R1), and flexible orifice (F1) were used in a cylindrical bubble column with 5 cm diameter and 30 cm height. From their experiments, the k_La were clearly affected by the gas velocity (V_g) in the bubble column which was contrasted with the results of the membrane reactor. The k_La of the membrane system was superior especially at the low gas velocity with 20 units of membranes. As mentioned earlier, the significant parameter affected k_La of membrane system was the numbers of unit membrane module while the control variable for the bubble column was superficial velocity which was not flexible. The increase of the amounts of membranes would intensely rise the k_La to above the conventional bubble column; the modification was easier comparing to the adaption of gas velocity in bubble columns. Hence, this membrane contactor had an advantage to bubble column not only in term of its adaptability but also its mass transfer performance.
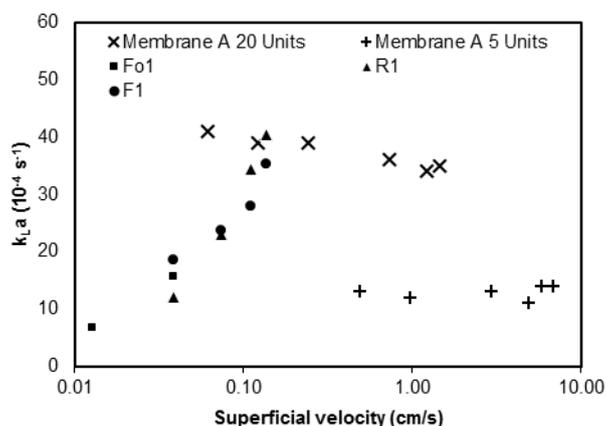

Fig. 7.   Comparison of the k_La between the membrane contactor and the cylindrical bubble column [20] in the different gas superficial velocity.

3.4. The Empirical Correlation for Aeration Process of the Hollow Fiber Membrane

The previous part indicated that the k_La from membrane contactor was the function of the liquid volumetric flow rate, the numbers of membrane tube and the surface type of membrane described by the contact angle (θ). With the purpose of further usage and design, the correlation between these variables was developed in term of dimensionless groups since it has an advantage in representing the system phenomena. With this purpose, the Buckingham Pi theorem was applied [21]. The dependent variable for the system was the k_La

while the independent variables were liquid density (ρ_L), liquid viscosity (μ_L), membrane outside diameter (d_o), liquid velocity in membrane (V_l), contact angle of surface membrane (θ), numbers of membrane in system (n) and inside diameter of module (D_i). After the Buckingham Pi Theorem was applied the relationship of variables can be described as in Eq. (12).

$$\frac{k_L a V_L}{g} = f(\text{Re}_L, \phi, \theta) \tag{12}$$

In the equation, the $k_L a$ is formed the dimensionless group with V_l and g and is the function of liquid Reynolds number (Re_L), packing density ($\emptyset$) and contact angle of membrane surface (θ). The liquid Reynolds number and packing density are expressed in Eq. (13) and (14), respectively.

$$\text{Re}_L = \frac{\rho_L V_L d_i}{\mu_L} \tag{13}$$

$$\phi = n \left(\frac{d_o}{D_i} \right)^2 \tag{14}$$

The non-linear regression was applied to obtain the relations between each group and found that the relation was in form of exponential as showed in Eq. (15). Each parameter of the equation is expressed in Eq. (16)-(18).

$$\frac{k_L a V_L}{g} = K \, \text{Re}_L^a \, \phi^b \tag{15}$$

where;

$$K = 2.68 x 10^{-10} \left[3,500 - 111(\theta) + (\theta)^2 \right] \tag{16}$$

$$a = 1.29 + 0.0069(\theta) - 5.2 x 10^{-5}(\theta)^2 \tag{17}$$

$$b = 2.45 - 0.033(\theta) + 2.21 x 10^{-4}(\theta)^2 \tag{18}$$

From Eq. (15), the mass transfer coefficient dimensionless group is majorly depending on the liquid Reynold number and packing density as both variables were in exponential function to it. However, the contact angle plays an important role as it affects all parameters in Eq. (15) in form of parabolic function, Eq. (16)-(18). The optimum contact angle for the aeration process predicting with Eq. (15) is 98 degrees which is the hydrophobic surface membrane that was claimed to be better than hydrophilic [14]. For validation, this model accuracy was identified by comparing the calculated values from Eq. (15) and the experimental values at the same condition as depicted in Fig. 8. The figure indicates that the $k_L a$ value from the equation are consistent with the experimental results with the average deviation than $\pm$ 13.26 %.
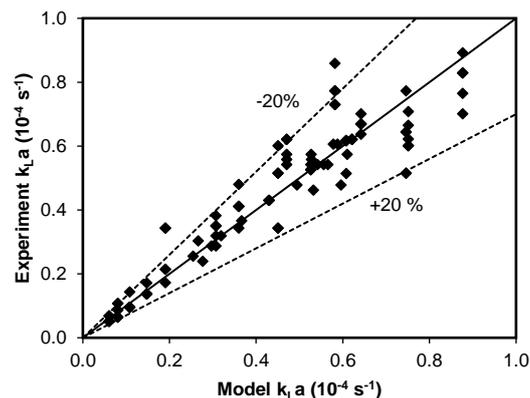
Fig. 8. Comparison of $k_L a$ from the experiment and the empirical correlation for aeration in hollow fiber membrane.

3.5. Application of Hollow Fiber Membrane: CO_2 Absorption

In this part, the $k_L a$ for CO_2 absorption and CO_2 removal efficiency were studied. The liquid phase was constantly circulated at 0.15 l/min for all the CO_2 experiment.
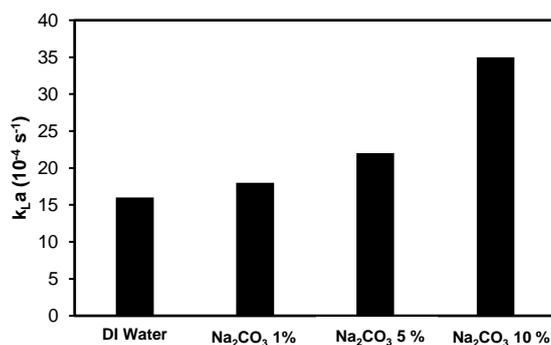

Fig. 9. Comparison of $k_L a$ between physical and chemical absorption.

Figure 9 illustrates the results of CO_2 absorption in different Na_2CO_3 concentration. The increase of Na_2CO_3 concentration rose the $k_L a$ value since CO_2 is the acid gas which can rapidly react with base solution of Na_2CO_3. Therefore, as can be seen in Fig. 10 which compares the CO_2 removal efficiency between at different concentrations of Na_2CO_3 for time up to 30 minutes, the treatment efficiency of CO_2 rose rapidly at time between 0-10 minutes especially for higher concentration of Na_2CO_3. The treatment efficiency subsequently went up slowly to approximate 80 percent for any concentration of Na_2CO_3 and 62 percent for DI water. For time between 0-3 minute, the higher concentration of Na_2CO_3 resulted in faster reaction and higher $k_L a$; the treatment efficiency for higher concentration was higher over this period. However, after 3 minute, the different of Na_2CO_3 concentration didn't have clearly effect on the treatment efficiency despite of their $k_L a$ value. This occurred due to the decrease of saturated concentration CO_2 when presenting of Na_2CO_3 particularly at high concentration [22]. The decrease of CO_2 saturated concentration reduced the mass transfer rate as C_L^* in Eq. (1) was decreased.
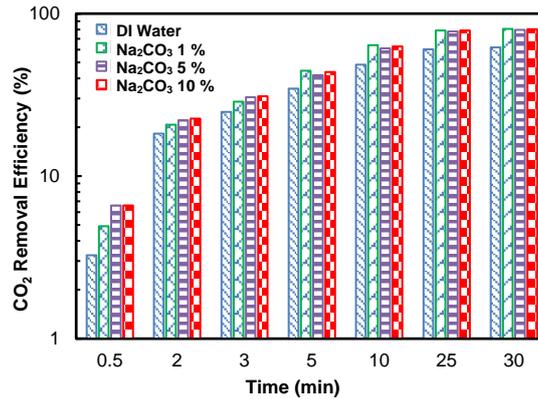
Fig. 10. Comparison of CO_2 removal efficiency between physical and chemical absorption.

Moreover, the prediction model for CO_2 absorption was developed following the methodology of the aeration empirical model. From the non-linear regression, the prediction equation for CO_2 absorption was found to be in term of exponential function with liquid Reynolds number (Re_L) and initial gas concentration ($C_{g,initial}$). The chemical absorption was also concerned by addition of Na_2CO_3 concentration term as expressed in Eq. (16). The accuracy of this equation is very satisfactory with the deviation less than ± 3.77 % as depicted in Fig. 11.

$$\frac{k_L a V_L}{g} = 1.03x10^{-11} C_{g,initial}^{0.92} Re_L^{1.26} + 4.21x10^{-6} C_{Na_2CO_3} \tag{16}$$
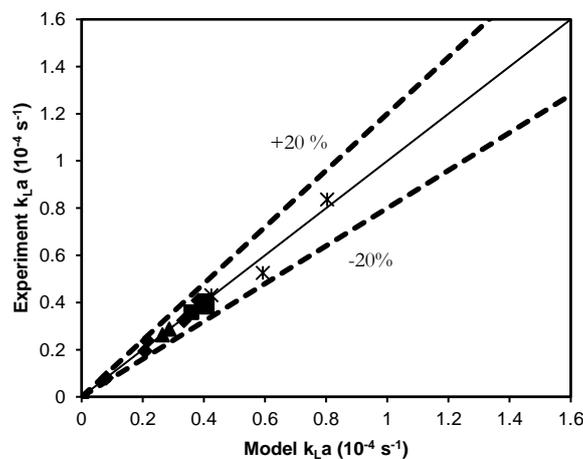

Fig. 11. Comparison of $k_L a$ from the experiment and the empirical correlation for CO_2 absorption in hollow fiber membrane.

4. Conclusion

This research investigated the overall mass transfer coefficient ($k_L a$) for three different types of microporous hollow fiber membrane which normally used in filtration process. These types of membranes are cheaper than the conventional membrane used in the absorption process. It was found that these membranes could be used for the aeration process and absorption of CO_2 effortlessly. The membranes also had an advantage over a bubble column as it could increase its mass transfer rate flexibly by increasing the number of membranes that resulted in rising its interfacial area significantly. Moreover, the effects of each operating parameter were determined. The consistent results with the commercial absorption membrane were obtained. For the same number of membrane tube, the membrane with the largest porous diameter yielded the highest $k_L a$ value since it provided greater contact area. The contact angle also played an important role regarding

their operating surfaces. The hydrophobic surface trended to have higher $k_L a$ value for aeration process since the operating surface was dry when operating with water. The $k_L a$ value was also affected with the increase of the liquid flow rate but rarely affected on the gas flow rate because the mass transfer rate was limiting at the liquid phase. The membrane could also applied to absorb CO_2 gas into liquid phase. It was confirmed that the carbon dioxide treatment efficiency up to approximate 80 percent could be reached within 25 minute with the assistance of Na_2CO_3 base solution. The empirical equations for prediction of the mass transfer coefficient for both aeration and CO_2 absorption were proposed with the acceptable accuracy.

Nomenclature

a	Specific Interfacial Area (m^{-1})
c_L	Dissolved Oxygen Concentration Saturation at Specific Time (mg/l)
c^*	Maximum Dissolved Oxygen Concentration Saturation (mg/l)
$c_{g,initial}$	Initial Gas Concentration (mg/l)
$c_{g,t}$	Gas Concentration at Specific Time (mg/l)
d_i	Inside Diameter of Hollow Fiber Membrane (m)
d_o	Outside Diameter of Hollow Fiber Membrane (m)
D_i	Inside Diameter of Module (m)
D_{iL}	The Diffusion Coefficient (m^2/s)
ESEM	Environment Scanning Electron Microscope
g	Gravitational Acceleration (m/s^2)
k_H	Henry 's Law Constant (L·atm/mol)
$k_L a$	Overall Mass Transfer Coefficient (1/s)
k_L	Liquid-Side Mass Transfer Coefficient (m/s)
L	Effective Length of The Membrane Module (m)
n	Number of Hollow Fiber Membrane
Q_g	Gas Flow Rate (l/min)
Q_l	Liquid Flow Rate (l/min)
Re	Reynolds Number
Sh	Sherwood Number
t	Time (s)
V_g	Gas Velocity (m/s)
V_l	Liquid Velocity (m/s)

Greek letters

ϱ_l	Liquid Density (kg/m^3)
μ_l	Viscosity of Liquid (Pa·s)
Ø	Membrane Module Packing Density
θ	Contact Angle of Membrane Surface $(^O)$

Acknowledgement

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