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Research Article
CO_2 Absorption from its Mixture Through Super-hydrophobic Membrane Contactor

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Abstract
Objective: The aim of this study is to evaluate the performance of super-hydrophobic hollow fiber membrane contactors to absorb CO_2 from its mixture with N_2 or CH_4 using 5 wt% diethanolamine (DEA) solution as absorbent. **Methodology:** During the experiment the absorbent flowed through lumen fibers, whilst the feed gas flowed through the shell side of the membrane contactor. **Results:** The experimental results demonstrated that the mass transfer coefficients, the fluxes and the amount of CO_2 absorbed increased with the absorbent and feed gas flow rates for CO_2-N_2 and CO_2-CH_4 feed gas system. The CO_2 absorption efficiency for both feed gas increased with the absorbent flow rate. The CO_2 absorption efficiency increased with feed gas flow rate for CO_2-N_2 system, but decreased for CO_2-CH_4 system. **Conclusion:** The overall mass transfer coefficient and the flux decreased with the number of fibers in the contactor, whilst, the amount of CO_2 absorbed and the CO_2 absorption efficiency increased.

Key words: Flux, mass transfer coefficient, membrane contactor, super-hydrophobic

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Carbon dioxide (CO_2) is one of the most important greenhouse gases produced by industries such as natural gas and power plant industries[1]. There are many methods that can be used to capture CO_2 either form natural gas or from flue gas such as absorption through chemical and physical absorbents[2], absorption through solid surface[3] and absorption through membrane contactor[4]. Chemical absorption in absorption column was the most established method for decades especially in petroleum and natural gas industries. However, this method has several disadvantages such as large space, high capital cost and operational problems e.g., liquid channeling, flooding, entrainment and foaming. Therefore, there is a need to develop new alternative technology to enhance the efficiency of absorption process and to reduce the effect of disadvantages[5]. Hollow fiber membrane contactor is a promising alternative and has been applied in many processes such as ammonia removal from wastewater and CO_2 absorption processes[6,7]. Hollow fiber membrane contactor contains two channels in which absorbent liquid and gas mixture are in contact with each other without getting mixed, so an increase in the fluid velocity through the channels will not lead to the common operational problems in tower such as flooding, entrainment and foaming[8]. Therefore, gas–liquid hollow fiber membrane contactors can be an alternative substitute technology with high removal efficiency for absorption of pollutant[9].

The CO_2 absorption through the membrane contactor is generally focused on the use of porous membranes, which are suffered if the membrane pores are wetted by the solvent[10]. The wetting of membrane pores significantly affected the mass transfer coefficients as the resistance of membrane phase increasing[11,12]. It was reported that the membrane wetting can cause a significant drop of CO_2 flux for both physical and chemical absorptions up to 90% when water used as the absorbent[13]. In this case, an assembly of ultrathin films incorporating carbonic anhydrase can be applied to microporous hydrophobic membrane to reduce pore wetting[14]. Therefore, the selection of the liquid absorbents is an important measures to prevent the wetting of the membrane[15]. In addition, the high temperature of absorbent can reduce the mass transfer coefficient in the membrane contactor[16].

The other important element of the membrane contactor is membrane material, which should have important properties such as high hydrophobicity, high surface porosity, low mass transfer resistance as well as resistance to various chemical liquids[17]. The high hydrophobicity property is needed to prevent the wetting of membrane pores that can increase the overall mass transfer coefficient and therefore, the choice of membrane material is critical to the efficiency of the process[18]. There are many membrane materials that can be utilized as contactor for gas-liquid operation such as popypropylene (PP)[9], polyvynilidinefluoride (PVDF)[19] and polyvynilchloride (PVC)[20]. The membrane can be incorporated with other material such as graphene nano sheets[21] or prepared as super-hydrophobic membrane[22] to improve the hydrophobicity of the membrane. This study aims to evaluate the performance of super hydrophobic hollow fiber membrane contactor to absorb CO_2 using water and diethanolamine (DEA) solution as absorbents. The flow rates of absorbents as well as the number of fibers in the membrane contactor will be observed to see their effects on CO_2 absorption processes in the contactor[23].

MATERIALS AND METHODS

Figure 1 shows the schematic diagram of CO_2 absorption from its mixture with N_2 or CH_4 using a membrane contactor, which has already reported in the literature[7]. The super hydrophobic hollow fiber membrane contactors used were supplied by PT GDP Filter Bandung. The feed gas (CO_2:N_2 = 13:87 and CO_2:CH_4 = 36:64) and DEA were purchased from BOC Gases and Merck, respectively. The super-hydrophobic fiber membrane used is polypropylene-based with outer and inner diameter of about 525 and 235 μm, respectively. Hollow fiber membrane contactors used for CO_2-N_2 mixture were 6 and 25 cm in diameter and length, respectively, which consist of 1000, 2000 and 5000 fibers (FA1000, FA2000 and FA5000). Meanwhile, hollow fiber membrane contactors used for CO_2-CH_4 mixture were 8 and 25 cm in diameter and length, respectively, which consist of 2000, 4000 and 8000 fibers (FB2000, FB4000 and FB8000). During the experiment, the absorbents of DEA solution (5% v in water) flowed through the lumen side of the membrane fiber, whilst the feed flowed through the shell side of the membrane contactor. The absorbents and gas flow rates were measured using liquid flow meter Krohne and mass flow meter Sierra Top Trak Instruments, respectively. Meanwhile, inlet and outlet gas compositions were measured using gas chromatography Bruker Scion 436-GC.

The overall mass transfer coefficient, K_{OVL}, will be calculated by Wang *et al.*[24]:

$$K_{OVL} = \left(\frac{Q_{Gin}}{A_m}\right)\ln\left(\frac{C_{in}}{C_{out}}\right) \quad (1)$$

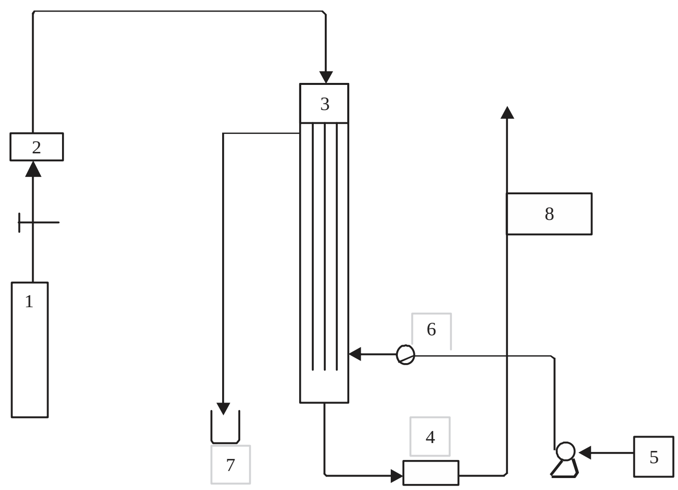

Fig. 1: Schematic diagram of experimental set up, 1: Feed gas, 2: Mass flow meter, 3: Super-hydrophobic hollow fiber membrane contactor, 4: Mass flow meter, 5: Absorbent reservoir, 6: Flow meter, 7: Absorbent reservoir, 8: Gas chromatography

where, Q_{Gin}, A_m, C_{in} and C_{out} are inlet gas flow rate, membrane surface area and CO_2 concentration in the inlet and outlet gas, respectively. Meanwhile, CO_2 flux through the membrane contactor, J and CO_2 absorption efficiency and R (%) were calculated by:

$$J = (Q_{iGin} - Q_{iGout}) \times \frac{RT}{Am} \quad (2)$$

$$R(\%) = \frac{100(Q_{iGin} - Q_{iGout})}{Q_{iGin}} \quad (3)$$

where, $Q_{iGin,}$ Q_{iGout}, T and R are the CO_2 flow rates in the inlet and outlet gas, temperature and gas constant, respectively.

RESULTS AND DISCUSSION

Based on the experiments, there are three mass transfer resistances in the process of CO_2 absorption through the hollow fiber membrane contactors, namely: Mass transfer resistance in the gas phase in the shell side of the membrane contactors; mass transfer resistance in the pores of the membrane fiber and mass transfer resistance in the absorbents phase in the lumen fibers. Therefore, the equation of mass transfer coefficient in the membrane contactors can be written as:

$$\frac{1}{K_{OV}} = \frac{H}{k_G} + \frac{H}{k_m} + \frac{1}{k_L} \quad (4)$$

where, K_{OV}, k_G, k_m, k_L and H are overall mass transfer coefficient, mass transfer coefficient in gas, membrane and liquid phases and Henry's constant, respectively.

The effect of the absorbent flow rate and the number of fiber in the membrane contactors at the same feed gas flow rate on the overall mass transfer coefficient is shown in Fig. 2. Figure 2 shows that, for the CO_2-N_2 and CO_2-CH_4 system, the mass transfer coefficient increase with increasing the absorbent flow rate in the lumen fiber in the membrane contactor. The mass transfer resistance in the hollow fiber membrane contactor is usually dominated in the liquid phase[25]. An increase in the absorbent flow rate will increase the turbulence in the liquid boundary layer that will reduce the mass transfer resistance in the liquid phase and based on Eq. 4, will increase the overall mass transfer coefficient[26,27]. The increase in the mass transfer coefficient with the absorbent flow rates indicated that the membrane was not wetted by the absorbent[24]. The increase in the overall mass transfer coefficient with increasing solvent flow rate in the membrane contactors also reported by Kim and Yang[28]. In this study, the overall mass transfer coefficient, K_L, increase from 0.00034-0.00039 and 0.00023-0.00027 cm sec^{-1} for CO_2-N_2 and CO_2-CH_4 system, respectively, if the absorbent flow rate increase from 100-500 mL min^{-1}. Meanwhile, Kim and Yang[28] reported that the overall mass transfer coefficient, K_L, increase from 0.0014-0.0025 cm sec^{-1} if the absorbent flow rate increase from 10-130 mL min^{-1} for CO_2-N_2 (40:60) system using amine solution as absorbent.

The numbers of fibers in the contactors have the same effect for both feed gas on the overall mass transfer coefficient

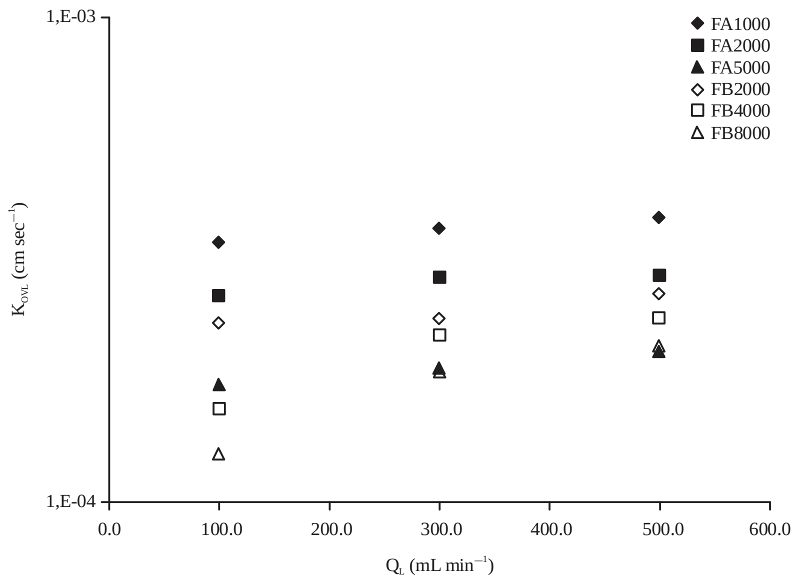

Fig. 2: Effects of absorbent flow rate, Q_L, on the overall mass transfer coefficient, K_{OVL}, at the membrane contactor consists of 1000, 2000 and 5000 fibers for CO_2-N_2 system and 2000, 4000 and 8000 fibers for CO_2-CH_4 system

at the same absorbent and feed gas flow rate. The increase in the number of fibers in the contactor will reduce the absorbent flow rate in the individual fiber at the same overall absorbent flow rate, which will reduce the overall mass transfer coefficient[29]. On the other hand, the increase in the number of fibers in the contactor will increase the contact area for the mass transfer, which led to the increase in the overall mass transfer coefficient. Figure 2 shows that the overall mass transfer coefficient for both feed gas decrease with increasing the number of fibers in the contactor at the same absorbent and feed gas flow rate, indicating that the effect of the absorbent flow rate more dominant than the effect of the surface area for contact. Figure 2 also shows that the overall mass transfer coefficient for CO_2-N_2 system is higher than for CO_2-CH_4 system due to the decrease in the CO_2 inlet concentration. The similar phenomena also shown by Wang *et al.*[24], who reported that the overall mass transfer coefficient decrease with increasing the CO_2 concentration in the feed gas using 2 M Na_2CO_3 aqueous solution as absorbent.

Similar to the mass transfer coefficient, the flux of CO_2 through the membrane fibers increase with increasing the absorbent flow rate in the membrane contactor due to the turbulence and surface renewal effects[26,27,30] as shown in Fig. 3. Several researchers also demonstrated the same trend where the flux increases with increasing solvent flow rate[16,31]. In this study, the flux, J, increase from 8.3×10^{-6} to 8.8×10^{-6} mmol cm^{-2} sec^{-1} and 2.9×10^{-6} to 3.2×10^{-6} mmol cm^{-2} sec^{-1} for CO_2-N_2 and CO_2-CH_4 system, respectively, if the absorbent flow rates increase from 100-500 mL min^{-1}. Gong *et al.*[31] reported that the flux of CO_2 increase from 1.6-1.9 mmol m^{-2} sec^{-1} if the absorbent of 1 wt% MEA and 9 wt% MDEA solution flow rate increase from 20-50 mL min^{-1}. Meanwhile, Yan *et al.*[16] reported that the flux of CO_2 increase from 2.2-3.0 mol m^{-2} h^{-1} if the absorbent of 1 M Potassium Glycinate (PG) velocity increase from 0.21-0.56 m sec^{-1}.

Figure 3 also shows that the flux of CO_2 through the membrane fibers for both feed gas, at the same absorbent and feed gas flow rate decrease with the increase in the number of fibers in the contactor, indicating that the effect of the absorbent flow rate more dominant than the effect of the surface area for contact. As for the overall mass transfer coefficient, the flux for CO_2-N_2 system are higher than for CO_2-CH_4 system due to the decrease of CO_2 concentration in the inlet gas[24].

Figure 4 and 5 show the effect of the absorbent flow rate on the CO_2 absorption efficiency, R (%) and the amount of CO_2 absorbed for both feed gas at the various numbers of fibers in the membrane contactors. The CO_2 absorption efficiency and the amount of CO_2 absorbed increase with increasing the absorbent flow rate in the membrane contactor due to increasing turbulence the absorbent boundary layer[24,26,27,30]. The CO_2 absorption efficiency and the amount of CO_2 absorbed also increase with the increase in the number of fibers in the membrane contactor due to more contact surface area for absorption. The same results also reported by Gong *et al.*[31] and Kim and Yang[28]. In this study, the CO_2

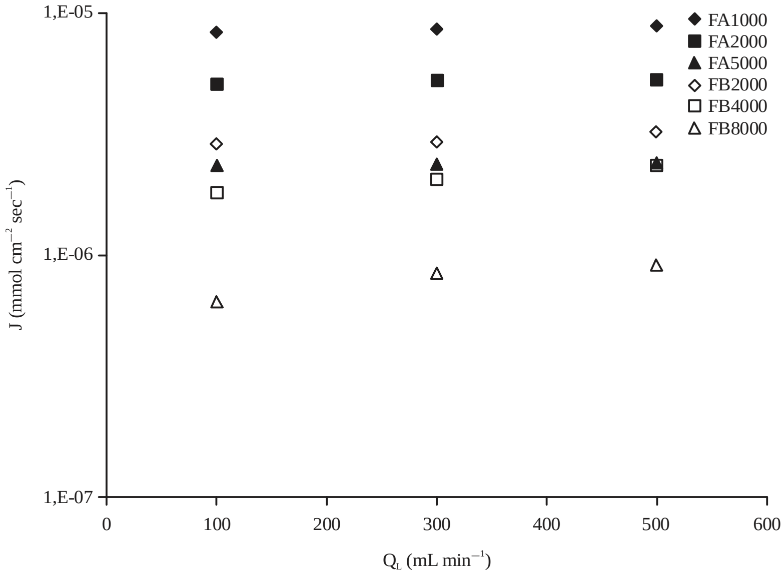

Fig. 3: Effects of absorbent flow rate, Q_L, on the flux, J, at the membrane contactor consists of 1000, 2000 and 5000 fibers for CO_2-N_2 system and 2000, 4000 and 8000 fibers for CO_2-CH_4 system

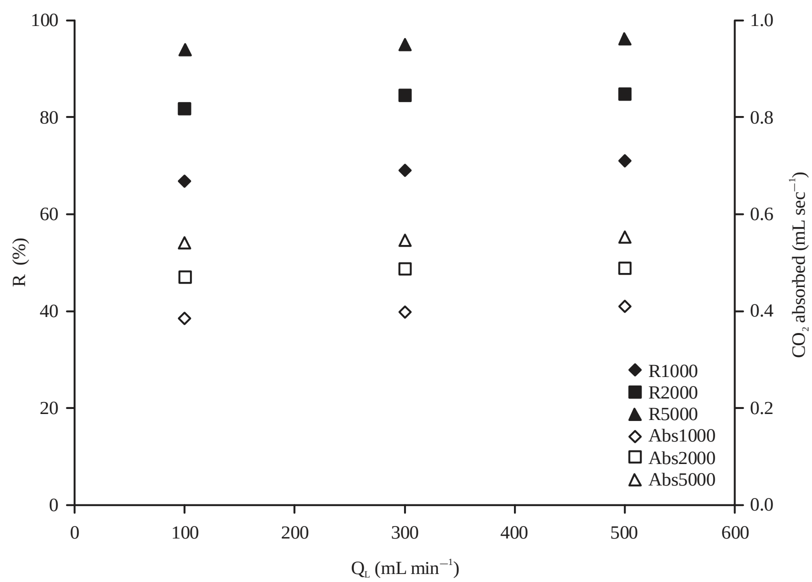

Fig. 4: Effects of absorbent flow rate, Q_L, on the absorption efficiency, R (%) and amount of CO_2 absorbed at the membrane contactor consists of 1000, 2000 and 5000 fibers for CO_2-N_2 system

absorption efficiency, R (%), increases from 93.9-96.2 and 54.2-81.4% for CO_2-N_2 and CO_2-CH_4 system, respectively, if the absorbent flow rates increase from 100-500 mL min^{-1}. Gong *et al*.[31] showed that the CO_2 absorption efficiency increase from 65-85% if the absorbent of 1 wt% MEA+9 wt% MDEA solution increase from 20-50 mL min^{-1}. Meanwhile, Kim and Yang[28] showed that the CO_2 absorption efficiency increases from 85-99% if the absorbent of 4 wt% MEA solution increase from 15-115 mL min^{-1}.

The effect of the feed gas flow rate and the number of fibers in the membrane contactor at the same absorbent flow rate on the overall mass transfer coefficients is shown in Fig. 6. Figure 6 shows that, for the CO_2-N_2 and CO_2-CH_4 system, the mass transfer coefficient increase with increasing the feed gas flow rate in the shell side of the membrane contactor. The increase of the feed gas flow rate can provide more CO_2 to be absorbed by the DEA solution which led to an increase in the overall mass transfer coefficients[32]. The same trend also

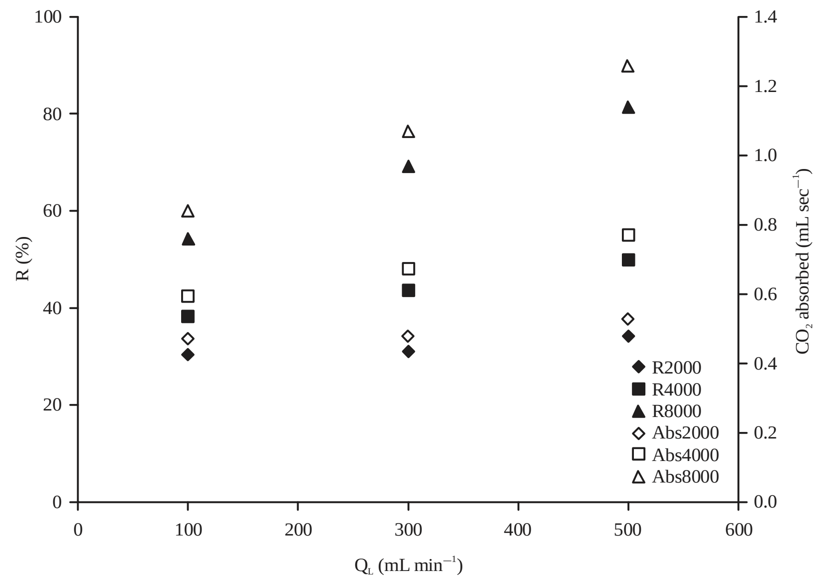

Fig. 5: Effects of absorbent flow rate, Q_L, on the absorption efficiency, R (%) and amount of CO_2 absorbed at the membrane contactor consists of 2000, 4000 and 8000 fibers for CO_2-CH_4 system

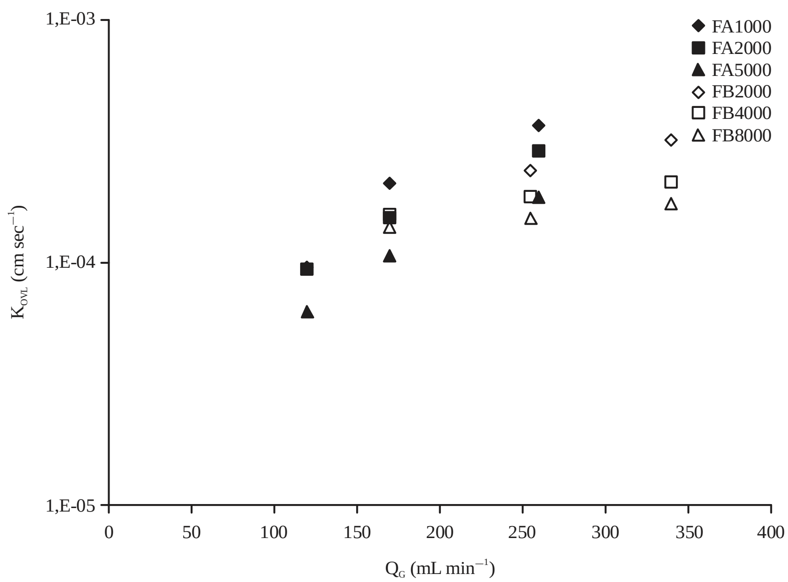

Fig. 6: Effects of feed gas flow rate, Q_G, on the overall mass transfer coefficient, K_{OVL}, at the membrane contactor consists of 1000, 2000 and 5000 fibers for CO_2-N_2 system and 2000, 4000 and 8000 fibers for CO_2-CH_4 system

reported by Wang *et al.*[32] using 2M DEA solution as absorbent for CO_2-N_2 system (20/80). In this study, the overall mass transfer coefficients, K_L, increase from 0.00009-0.00037 cm sec^{-1} and 0.00023-0.00032 cm sec^{-1} for CO_2-N_2 and CO_2-CH_4 system, respectively, if the feed gas flow rates increase from 120-260 mL min^{-1} and from 170-340 mL min^{-1}, respectively. Meanwhile, Wang *et al.*[32] reported that the overall mass transfer coefficients increase from 0.00016-0.00025 m sec^{-1} if the feed gas velocities increase from 0.03-0.09 m sec^{-1}.

The numbers of fibers in the contactors have the same effect for both feed gas on the overall mass transfer coefficient at the same absorbent and feed gas flow rates. The increase in the number of fibers in the contactor will reduce the residence time of the gas in the membrane contactor, which led to the decrease in the overall mass transfer coefficient. On the other hand, the increase in the number of fibers in the contactor will increase the contact area for the mass transfer, which led to the increase in the overall mass transfer coefficient. Figure 5 shows that the overall mass transfer

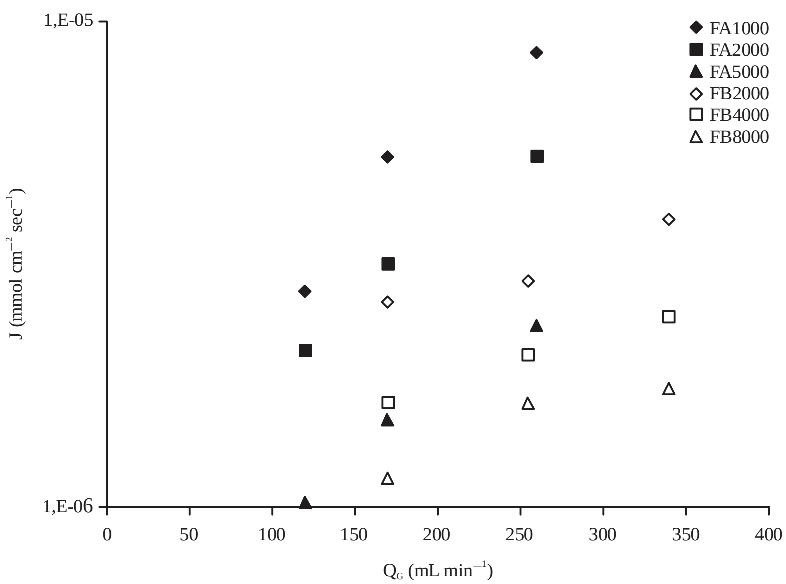

Fig. 7: Effects of feed gas flow rate, Q_G, on the flux, J, at the membrane contactor consists of 1000, 2000 and 5000 fibers for CO_2-N_2 system and 2000, 4000 and 8000 fibers for CO_2-CH_4 system

coefficients for both feed gas decrease with increasing the number of fibers in the contactor at the same absorbent and feed gas flow rates, indicating that the effect of the feed gas flow rate more dominant than the effect of the surface area for contact.

The impact of the feed gas flow rate and the number of fibers in the membrane contactor at the same absorbent flow rate on the flux is demonstrated in Fig. 7. Similar to the overall mass transfer coefficient, the flux increase with increasing the feed gas flow rate in the shell side of the membrane contactor due to more CO_2 that can be absorbed by the absorbent solution[16,32]. Similar result was also shown by Keshavarz *et al.*[12] and Zhang *et al.*[33]. In this study, the flux of CO_2, J, increase from 2.7×10^{-6} to 8.8×10^{-6} mmol cm^{-2} sec^{-1} and 2.6×10^{-6} to 3.9×10^{-6} mmol cm^{-2} sec^{-1} for CO_2-N_2 and CO_2-CH_4 system, respectively, if the feed gas flow rates increase 120-260 and 170-340 mL min^{-1}, respectively. Meanwhile, Keshavarz *et al.*[12] and Zhang *et al.*[33] demonstrated that the flux of CO_2 increase from 0.0003-0.0008 mol m^{-2} sec^{-1} if the feed gas flow rate increase from 0.03-0.09 m sec^{-1} for CO_2-N_2 system (20/80) using 2 M DEA solution as absorbent. Figure 7 also shows that the flux for both feed gas decrease with increasing the number of fibers in the contactor, at the same absorbent and feed gas flow rates, indicating that the effect of the feed gas flow rate more dominant than the effect of the surface area for contact.

Figure 8 and 9 show the effect of the feed gas flow rate on the CO_2 absorption efficiency, R (%) and amount of CO_2 absorbed at the various numbers of fibers in the membrane contactors. The amount of CO_2 absorbed for both feed gas increase with increasing the feed gas flow rate in the membrane contactor due to increasing turbulence the absorbents boundary layer[24,26,27,30] as shown in Fig. 8 and 9. The amount of CO_2 absorbed also increase with the increase in the number of fibers in the membrane contactor due to more contact surface area for absorption. The same results also reported by Yan *et al.*[16] using 1 M PG or 1 M MEA as absorbents. However, both feed gas system show the different phenomena for the CO_2 absorption efficiency, R (%). The CO_2 absorption efficiency increases with increasing feed gas flow rate for CO_2-N_2 system and decreases with increasing feed gas flow rate for CO_2-CH_4 system. The decrease in the CO_2 absorption efficiency with increasing gas flow rate is also shown by Yan *et al.*[16]. In this study, the CO_2 absorption efficiency for CO_2-CH_4 system decrease from 72.7-64.0% if the gas flow rate increase from 170-340 mL min^{-1} using 5% DEA solution as absorbent. Meanwhile, Yan *et al.*[16] showed that the CO_2 absorption efficiency for CO_2-N_2-O_2 system decrease from 94-65 and 89-54% if the gas velocity increase from 0.21-0.56 m sec^{-1} using 1 M PEG and 1 M MEA as absorbents, respectively.

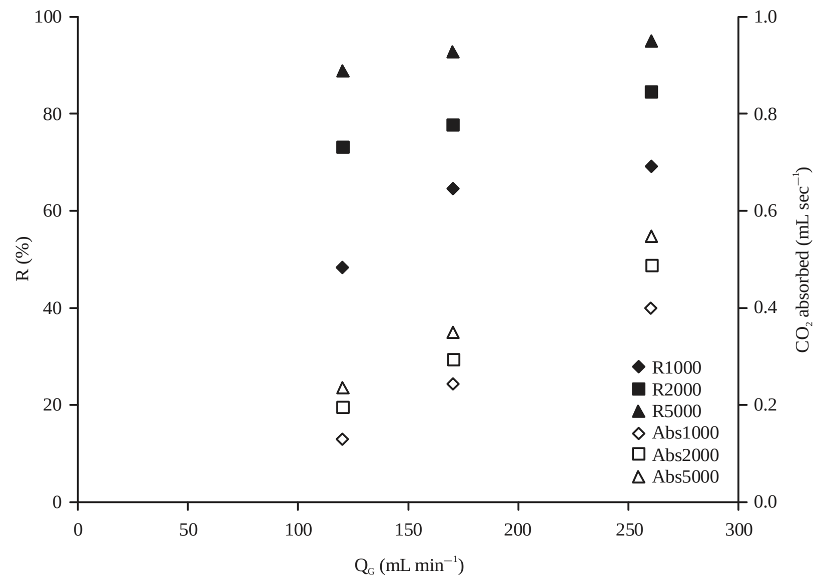

Fig. 8: Effects of feed gas flow rate, Q_G, on the absorption efficiency, R (%) and amount of CO_2 absorbed at the membrane contactor consists of 1000, 2000 and 5000 fibers for CO_2-N_2 system

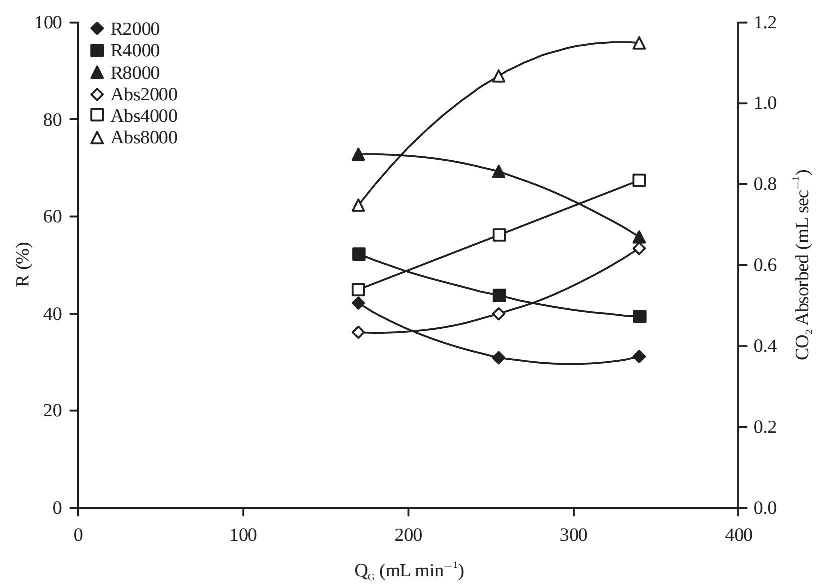

Fig. 9: Effects of feed gas flow rate, Q_G, on the absorption efficiency, R (%) and amount of CO_2 absorbed at the membrane contactor consists of 2000, 4000 and 8000 fibers for CO_2-CH_4 system

CONCLUSION

High hydrophobicity is one of the important properties of the membrane for gas-liquid contact process, which is required to avoid pores membrane wetting so that can increase the overall mass transfer coefficient. This study utilized polypropylene super hydrophobic hollow fiber membrane contactor to absorb CO_2 from its mixture with N_2 or CH_4 using DEA solution (5% vol in water) as absorbent. In the experiments, the absorbent flowed through the lumen of fibers, while the feed gas flowed through the shell side of the contactor. The experimental results showed that the mass transfer coefficients, the fluxes and the amount of CO_2 absorbed increased with increasing the absorbent and feed gas flow rates. The CO_2 absorption efficiency for both feed gas increased with increasing the absorbent flow rate. However, the increase in the feed gas flow rate gives different effect to the CO_2 absorption efficiency for CO_2-N_2 and CO_2-CH_4 system.

The increased in the feed gas flow rate will increase the CO_2 absorption efficiency for CO_2-N_2 system, but will decrease the CO_2 absorption efficiency for CO_2-CH_4 system. The numbers of fibers in the contactors gave the same effects for both feed gas system in terms of the overall mass transfer coefficient, the flux, the amount of CO_2 absorbed and the CO_2 absorption efficiency. The overall mass transfer coefficient and the flux decreased with the increase in the number of fibers in the contactor, whilst, the amount of CO_2 absorbed and the CO_2 absorption efficiency increased.

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