International Journal of Technology (2017) 8: 1429-1435
ISSN 2086-9614

EFFECTS OF ABSORBENT FLOW RATE ON CO_2 ABSORPTION THROUGH A SUPER HYDROPHOBIC HOLLOW FIBER MEMBRANE CONTACTOR

Sutrasno Kartohardjono[1,*], Angeline Paramitha[1], Aulia Andika Putri[1], Ryan Andriant[1]

[1]*Process Intensification Laboratory, Department of Chemical Engineering, Faculty of Engineering, Universitas Indonesia, Kampus UI Depok, Depok 16424, Indonesia*

(Received: January 2017 / Revised: May 2017 / Accepted: November 2017)

ABSTRACT

The aim of this study is to evaluate the effects of absorbent flow rate on CO_2 absorption through a super hydrophobic hollow fiber contactor. The absorbent used in this study was a physical absorbent, namely a polyethyleneglycol-300 (PEG-300) solution. Meanwhile, the feed gases used in the experiments were pure CO_2 and a mixture of 30% CO_2 and 70% CH_4. Gas absorption using a physical absorbent provides various benefits; for example, it can produce sufficiently high selectivity towards CO_2 and it is less corrosive than chemical solvents. Three super hydrophobic hollow fiber contactors, each 6 cm in diameter and 25 cm in lengthconsist of 1000, 3000 and 5000 fibers, respectively, were used in this study. The type ofsuper hydrophobic fiber membrane used was polypropylene-based, with an outer and inner diameter of about 525 and 235 µm, respectively. During the experiments, the absorbent was flowed through the lumen fibers, whilst the feed gas flowed through the shell side of the membrane contactors. The experimental results showed that the mass transfer coefficient, the flux, and the absorption efficiency increased, but the CO_2 loading decreased, with increasing absorbent flow rate in the membrane contactor. Meanwhile, it was found that an increase in the number of fibers in the membrane contactor, in general, will increase the absorption efficiency and the CO_2 loading, but will decrease the overall mass transfer coefficient and the flux.

*Keywords:*Absorption efficiency; Flux; Mass transfer coefficient; Physical absorbent; Polyethyleneglycol-300

1. INTRODUCTION

Natural gas, in addition to containing hydrocarbon compounds, contain impurities, such as CO_2, H_2S, N_2 and water. CO_2 is the impurity that will be the focus of this study. The presenceof CO can reduce the heating value of natural gas and can cause corrosion in the pipe lines; assuch, CO_2must be removed from natural gas. This is usually achieved through absorption in the absorption column or tower, which has some disadvantages, such as entrainment, flooding, unloading and foaming (Yeon et al., 2005). One alternative process that can be used to avoid the disadvantages that occurs in columns or towers for gas-liquid contactis a membrane process (Freeman et al., 2014).Because of the importance of this CO_2 gas separation process, various alternative shave been created through the modification of membrane technology, for example hydrophobic hollow fiber membrane contactors(Rajabzadeh et al., 2009). In addition to low operating costs, the hollow fiber membrane gas-liquid contactor has a larger contact area per unit volume than conventional gas-liquid contactors, such as column and tower contactors. By using hydrophobic membrane contactors,problems such as flooding, entertainment and

---

[*] Corresponding author's email: sutrasno@che.ui.ac.id, Tel: +62-21-7863516, Fax: +62-21-7863515
Permalink/DOI: https://dx.doi.org/10.14716/ijtech.v6i8.679

unloading, which occur when using conventional methods, can be eliminated as the gas and liquid flow ratescan be varied freely.

Many studies have been conducted both experimentally and theoretically on the absorption of CO_2 gas from various gas mixtures using various membrane and solvent materials in membrane contactors. These include: the use of polypropylene membranes and polytetraflorethylene membranes for CO_2 gas absorption(Tontiwachwuthikul & Chakma, 2006); gas-liquid contact via membrane contactor (Dindore et al., 2005); modeling the absorption of CO_2 through the membrane contactor either at low pressure or at high pressure(Dindore et al., 2004); and the selection of solvents for CO_2 removal processes through membrane contactors (Dindore et al.,2004). Based on the results of these studies, it is shown that it is very important to select the type of solvent and the membrane material carefully in order to maximize the efficiency of the process and to ensure that the solvent used to absorb CO_2 gas does not wet the membrane. The some of the results also suggest that the mass transfer resistance in the liquid phase is the dominant resistance to the mass transfer of CO_2 from the gas phase to the liquid phase.

This study will evaluate the influence of the solvent flow rate on the mass transfer coefficient, flux, and absorption efficiency in the CO_2 absorption process from its mixture with CH_4 or N_2 through the super hydrophobic membrane contactor using polyethylene glycol (PEG) solution as an absorbent. PEG is a physical solvent that can provide the advantagesof high selectivity and permeability to CO_2(Anda Lucia & Adiwar, 2011), requiring low power consumption for the solvent circulation, and being less corrosive than chemical solvents such as amines. Physical solvents such as PEG have been favored in the absorption process because of their natural affinity for acid gases (Daveet al., 2016).

2. MATERIALS AND METHODS

A schematic diagram of the experimental set up used in this study is shown in Figure 1, which has been reported elsewhere (Kartohardjono et al., 2016; Kartohardjono et al., 2017). The super hydrophobic hollow fiber membrane contactors used were supplied by PT GDP Filter Bandung. There were three contactors, each with a diameter of 6 cm and a length of 25 cmconsisting of 1000, 3000 and 500 fibers, respectively. The super-hydrophobic fiber polypropylene-based membrane used had an outer and inner diameter of 525 and 235 μm, respectively. Feed gas containing pure CO_2 or a mixture of CO_2 and CH_4 (36:64) was sent to the membrane contactor in the shell side, whilst PEG solution (5% vol.) was pumped to the membrane contactor in the lumen fiber. The feed gas and absorbent flow rates were measured using Sierra Top Trak Instrumentsmass flow meter and a Krohne liquid flow meter, respectively. Meanwhile, a gas chromatography instrument, the Bruker Scion 436-GC, was used to measure inlet and outlet gas compositions.

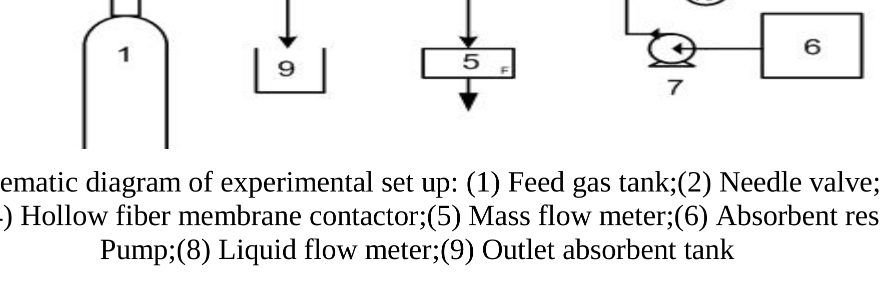

Figure 1 Schematic diagram of experimental set up: (1) Feed gas tank;(2) Needle valve;(3) Mass flow meter;(4) Hollow fiber membrane contactor;(5) Mass flow meter;(6) Absorbent reservoir;(7) Pump;(8) Liquid flow meter;(9) Outlet absorbent tank

The overall mass transfer coefficient, K_{OVL}, and CO_2 absorption efficiency, $\%R$, were calculated using the following equations (Wang et al., 2004):

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

$$\%R = \frac{C_{in} - C_{out}}{C_{in}} x100\% \tag{2}$$

Meanwhile, the CO_2 flux through the membrane contactor, J, (Kartohardjono et al., 2017) and acid loading were calculated as follows:

$$J = (Q_{iGin} - Q_{iGout}) * RT/Am \tag{3}$$

$$Acid\ Loading = \frac{mol\ CO2\ Absorbed}{mol\ PEG} \tag{4}$$

where Q_{Gin}, Q_{Gout}, A_m, C_{in} and C_{out} are the inlet and outlet gas flow rates, membrane surface area, and CO_2 concentrations in inlet and outlet gas, respectively. Meanwhile,R and T are the ideal gas constant and temperature, respectively.

3. RESULTS AND DISCUSSION

Figure 2 shows the effect of absorbent flow rate on the CO_2 overall mass transfer coefficient through the membrane contactors. As demonstrated in the figure, for both feed gases, the mass transfer coefficient increased with an increase in the absorbent flow rate in the lumen fiber of the contactor. This effect is due to increasing turbulence in the solvent boundary layer associated with the higher solvent flow rate whichcauses a reductionin resistance to CO_2 transfer through the membrane contactor (Franco et al., 2008). Similar trends were also found by Kim and Yang (2000) and (Scholes et al.,2012). In this study, the mass transfer coefficient increased from 1.6 to 4.5×10^{-5} $cm.s^{-1}$when the liquid flow rate increased from 100 to500 $ml.min^{-1}$. Meanwhile, Kim and Yang (2000) reported that the mass transfer coefficient increased from 1.1 to $2.2\times10^{-3}$$cm.s^{-1}$when the absorbent flow rate increased from 10 to 130 $ml.min^{-1}$,using 4 wt.% 2-amino-2-methyl-l-propanol (AMP) as an absorbent and feed gas containing a mixture of 40% $CO_2$and 60% N_2.

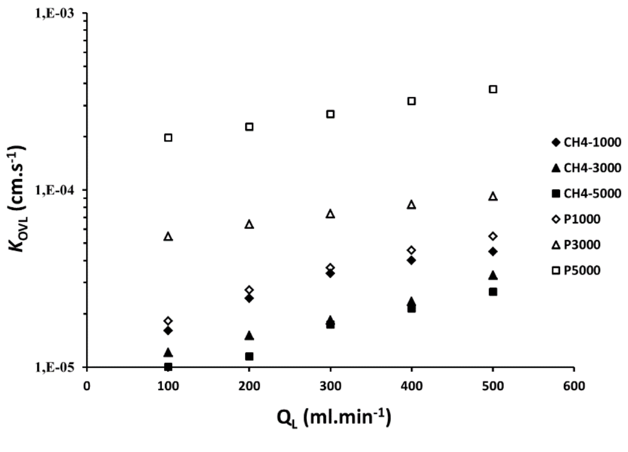

Figure 2 The CO_2 overall mass transfer coefficient, K_{OVL}, using 5 vol.% PEG solution as an absorbent on the lumen side and pure CO_2 (p1000, p3000 and p5000) or a mixture of CO_2-CH_4 (CH_4-1000, CH_4-3000 and CH_4-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers

Moreover, Scholes et al. (2012)demonstrated that the mass transfer coefficient increased from 3.8×10^{-5} to 1.5×10^{-4} $cm.s^{-1}$when the absorbent flow rate increased from 3 to 28 $ml.min^{-1}$,using 30 wt.% K_2CO_3 as an absorbent and feed gas containing a mixture of 10% $CO_2$and 90% N_2. Figure 2 also shows the effect of the number of fibers in the membrane contactor on the CO_2 overall mass transfer coefficient for both feed gases. The overall mass transfer coefficient increased with an increase in the amount of fibers in the membrane contactor for pure CO_2 feed gas, but decreased for CO_2-CH_4 feed gas. This finding indicates that the effect of surface area is more dominant in the pure CO_2 gas system, whilst the effect of absorbent flow rate is more dominant in the CO_2-CH_4 system.

Figure 3 shows the effect of absorbent flow rate on the CO_2 flux through the membrane contactors. Similarly, to the overall mass transfer coefficient, the CO_2 flux increased with an increase in the absorbent flow rate in the lumen fiber of the contactor for both feed gases, due to increasing turbulence in the solvent boundary layer. Similar trendswere also demonstrated by Yan et al. (2007) and Wang et al. (2005). In this study, the CO_2 flux increased from 7.3×10^{-7} to $4.52.1\times10^{-6}$ $mmol.cm^{-2}.s^{-1}$when the liquid flow rate increased from 100 to 500 $ml.min^{-1}$. Additionally, Yan et al. (2007) reported that the CO_2 flux increased from 0.8 to 1.0 $mol.m^{-2}.h^{-1}$when the absorbent velocity increased from 0.025 to 0.1 $m.s^{1}$, using a 3M MDEA solution as an absorbent and feed gas containing a 14 vol.% mixture of CO_2 with O_2 and N_2. Meanwhile, Wang et al. (2005) reported that the CO_2 flux increased from 3.0 to 8.0×10^{-4} $mol.m^{-2}.s^{-1}$when the absorbent flow rate increased from 0.03 to 0.09 $m.s^{-1}$, using a 2 M DEA solution as an absorbent and feed gas containing a mixture of 20% $CO_2$and 80% N_2. Similarly, to the overall mass transfer coefficient, the CO_2flux increased with an increase in the amount of fibers in the membrane contactor for pure CO_2 feed gas, due to the dominance of the surface area effect, but decreased for CO_2-CH_4 feed gas, due to the dominance of the absorbent flow rate effect.

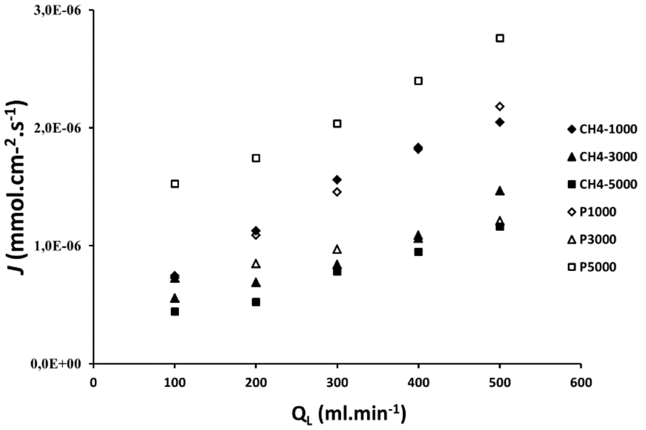

Figure 3 The CO_2 flux, *J*, using 5 wt.% PEG solution as an absorbent on the lumen side and pure CO_2 (p1000, p3000 and p5000) or a mixture of CO_2-CH_4 (CH_4-1000, CH_4-3000 and CH_4-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers

The effect of the absorbent flow rate on CO_2 absorption efficiency, %*R*, is shown in Figure 4. The CO_2 absorption efficiency increased with an increase in the absorbent flow rate in the membrane contactor, due to increasing turbulence in the absorbent boundary layer (Kartohardjono et al., 2017). Similar trendswere also reported in previous studies byKartohardjono et al. (2017) and Faiz and Marzouqi (2010). In this study, the CO_2 absorption efficiency, using a 5 vol.% PEG as an absorbent, increases from 8.1 to 14.6% for pure CO_2, and from 4.7 to 12.4% for the mixture of CO_2 and CH_4 (36:64),when the absorbent flow rate increases from 100 to 500 $ml.min^{-1}$. The study conducted byKartohardjono et al. (2017)found that CO_2absorption, using a 5 vol.% DEA solution as an absorbent, increases from 54.2−81.4% for a mixture of CO_2 and CH_4 (36:64) when the absorbent flow rate increases from 100 to 500ml.min^{-1}. Meanwhile, Faiz and Marzouqi (2010) reported that CO_2 absorption, using water as an absorbent, increases from 2.0 to 3.6% when the absorbent flow rate increases from 100 to 500 ml.min^{-1}. The CO_2 absorption efficiency also increases with increasing amounts of fiber in the membrane contactor; this is because the surface area for gas-liquid contact also increases.

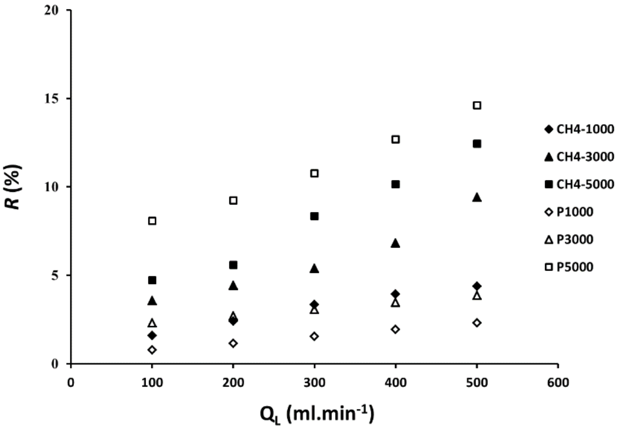

Figure 4 The CO_2 absorption efficiency, %R, using 5 wt.% PEG solution as an absorbent on the lumen side and pure CO_2 (p1000, p3000 and p5000) or a mixture of CO_2-CH_4 (CH_4-1000, CH_4-3000 and CH_4-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers

Figure 5 shows the effect of absorbent flow rate on the CO_2 loading in the membrane contactor. The CO_2 loading is defined as the amount of CO_2 absorbed per mole of absorbent introduced through the membrane contactor. For example, in the conventional column, the ideal CO_2 loading, using an amine solution as an absorbent, is 0.5 according to the equation below:

$$2RNH_2 + CO_2 + H_2O \xrightarrow{49\ oC} (RNH_2)_2H_2CO_3 \tag{5}$$

The CO_2 loading in this study decreases with increasing absorbent flow rate; this is due to the reduced absorbent residence time in the lumen fiber. Meanwhile, CO_2 loading increases with increasing amounts of fiber in the contactor,as a result of the increase in the surface area for the gas-liquid contact.

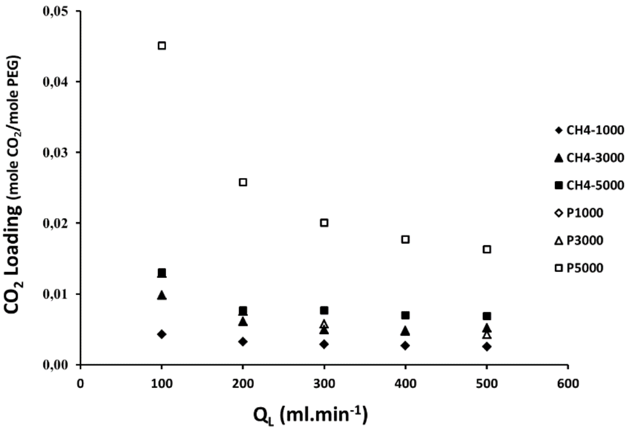

Figure 5 The CO_2 loading using 5 wt.% PEG solution as an absorbent on the lumen side and pure CO_2 (p1000, p3000 and p5000) or a mixture of CO_2-CH_4 (CH_4-1000, CH_4-3000 and CH_4-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers

4. CONCLUSION

The membrane-based gas-liquid contactor is a suitable alternative technology for CO_2 absorption from natural gas. Hydrophobicity is one of important properties of the membrane that prevents it being wetted by the absorbent solution. The super hydrophobic hollow fiber membrane contactors were utilized in this study to absorb CO_2 using 5 vol.% PEG as an absorbent. The experimental results showed that the mass transfer coefficient, flux and absorption efficiency increased, but CO_2 loading decreased, with increasing absorbent flow rate. The numbers of fibers in the contactor had different effects on mass transfer coefficient, flux and absorption efficiency. In general, the mass transfer coefficient and flux, at the same absorbent flow rate, decreased with increasing numbers of fibers in the membrane contactor. Meanwhile, the absorption efficiency and CO_2 loading, at the same absorbent flow rate, increased with increasing numbers of fiber in the membrane contactor.

5. ACKNOWLEDGEMENT

A part of this study was funded by the Directorate General of Higher Education Republic of Indonesia through Universitas Indonesia with contract No. 1178/UN2.R12/HKP.05.00/2016.

6. REFERENCES

Anda, L., Adiwar, 2011. The Effect of Molecular Weight of Polyethylene Glycol (PEG) on Acetate Cellulod Membranes on Separation Selectivity of CO_2/CH_4 Gases (*Efek Berat Molekul Polietilen Glikol (PEG) pada Membran Seluloda Asetat terhadap Selektifitas Pemisahan Gas* CO_2/CH_4. *Lembaran Publikasi Minyak dan Gas Bumi,* Volume 45(2), pp. 121−124 (in Bahasa)

Dave, A., Dave, M., Huang, Y., Rezvani, S., Hewitt, N., 2016. Process Design for CO_2 Absorption from Syngas using Physical Solvent DMEPEG. *International Journal of Greenhouse Gas Control,*Volume49, pp. 436−448

Dindore, V.Y., Brilman, D., Willem, F., Geuzebroek, F.H., Versteeg, G.F., 2004. Membrane–solvent Selection for CO_2 Removal using Membrane Gas–liquid Contactors. *Separation and Purification Technology,*Volume40(2), pp.133−145

Dindore, V.Y., Brilman, D., Willem, F., Versteeg, G.F., 2005. Hollow Fiber Membrane Contactor as a Sas–liquid Model Contactor. *Chemical Engineering Science,*Volume60(2), pp. 467−479

Dindore, V.Y., Brilman, D.W.F., Feron, P.H.M., Versteeg, G.F., 2004. CO_2 Absorption at Elevated Pressures using a Hollow Fiber Membrane Contactor. *Journal of Membrane Science,* Volume235(1), pp. 99−109

Faiz, R., Al-Marzouqi, M.,2010. CO_2 Removal from Natural Gas at High Pressure using Membrane Contactors: Model Validation and Membrane Parametric Studies. *Journal of Membrane Science,*Volume365(1), pp. 232−241

Franco, J., Demontigny, D., Kentish, S., Perera, J., Stevens, G.,2008. A Study of the Mass Transfer of CO_2 through Different Membrane Materials in the Membrane Gas Absorption Process. *Separation Science and Technology,* Volume43(2), pp. 225−244

Freeman, B., Hao, P., Baker, R., Kniep, J., Chen, E., Ding, J., Rochelle, G.T., 2014. Hybrid Membrane-absorption CO_2Capture Process. *Energy Procedia,* Volume63, pp. 605−613

Kartohardjono, S., Darmawan, R., Karyadi, M.F., Saksono, N.,2016. CO_2 Absorption through Super-hydrophobic Hollow Fiber Membrane Contactors. *Journal of Environmental Science and Technology,*Volume 9(2), pp. 214−219

Kartohardjono, S., Sembiring, K.A., Rexy, S.R., Ghasani, F., Saksono, N.,2017. CO_2Absorption from Its Mixture through Super-hydrophobic Membrane Contactor. *Journal of Environmental Science and Technology,* Volume10(1), pp. 25−34

Kim, Y.-S., Yang, S.-M.,2000. Absorption of Carbon Dioxide through Hollow Fiber Membranes using Various Aqueous Absorbents. *Separation and Purification Technology*,Volume21(1), pp. 101−109

Rajabzadeh, S., Yoshimoto, S., Teramoto, M., Al-Marzouqi, M., Matsuyama, H.,2009. CO_2Absorption by using PVDF Hollow Fiber Membrane Contactors with Various Membrane Structures. *Separation and Purification Technology,*Volume69(2), pp. 210−220

Scholes, C.A., Simioni, M., Qader, A., Stevens, G.W., Kentish, S.E.,2012. Membrane Gas–solvent Contactor Trials of CO_2Absorption from Syngas. *Chemical Engineering Journal,* Volume195, pp. 188−197

Tontiwachwuthikul, P., Chakma, A.,2006. Using Polypropylene and Polytetrafluoroethylene Membranes in a Membrane Contactor for CO_2 Absorption. *Journal of Membrane Science,* Volume277(1), pp. 99−107

Wang, D., Teo, W.K., Li, K.,2004. Selective Removal of Trace $H_2S$from Gas Streams Containing CO_2 using Hollow Fibre Membrane Modules/Contractors. *Separation and Purification Technology*,Volume35(2), pp. 125−131

Wang, R., Zhang, H.Y., Feron, P.H.M., Liang, D.T.,2005. Influence of Membrane Wetting on CO_2 Capture in Microporous Hollow Fiber Membrane Contactors. *Separation and Purification Technology*,Volume46(1), pp. 33−40

Yan, S.-P., Fang, M.-X., Zhang, W.-F., Wang, S.-Y., Xu, Z.-K., Luo, Z.-Y., Cen, K.-F.,2007. Experimental Study on the Separation of $CO_2$from Flue Gas using Hollow Fiber Membrane Contactors without Wetting. *Fuel Processing Technology,* Volume88(5), pp. 501−511

Yeon, S.-H., Lee, K.-S., Sea, B., Park, Y.-I., Lee, K.-H.,2005. Application of Pilot-scale Membrane Contactor Hybrid System for Removal of Carbon Dioxide from Flue Gas. *Journal of Membrane Science,* Volume257(1), pp. 156−160