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Effect of Feed Gas Flow Rate on CO₂ Absorption through Super Hydrophobic Hollow Fiber membrane Contactor

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Abstract. Carbon dioxide is pollutant in natural gas that could reduce the heating value of the natural gas and cause problem in transportation due to corrosive to the pipeline. This study aims to evaluate the effects of feed gas flow rate on CO₂ absorption through super hydrophobic hollow fiber contactor. Polyethyleneglycol-300 (PEG-300) solution was used as absorbent in this study, whilst the feed gas used in the experiment was a mixture of 30% CO₂ and 70% CH₄. There are three super hydrophobic hollow fiber contactors sized 6 cm and 25 cm in diameter and length used in this study, which consists of 1000, 3000 and 5000 fibers, respectively. The super hydrophobic fiber membrane used is polypropylene-based with outer and inner diameter of about 525 and 235 µm, respectively. In the experiments, the feed gas was sent through the shell side of the membrane contactor, whilst the absorbent solution was pumped through the lumen fibers. The experimental results showed that the mass transfer coefficient, flux, absorption efficiency for CO₂-N₂ system and CO₂ loading increased with the feed gas flow rate, but the absorption efficiency for CO₂-N₂ system decreased. The mass transfer coefficient and the flux, at the same feed gas flow rate, decreased with the number of fibers in the membrane contactor, but the CO₂ absorption efficiency and the CO₂ loading increased.

Keywords: Absorption efficiency; Flux; Mass transfer coefficient; Membrane contactor; Super hydrophobic.

1. Introduction
Consumption of natural gas in the world reached 100 trillion cubic feet and is estimated to be 160 trillion cubic feet in 2035 [1]. Meanwhile, the consumption of natural gas as energy in Indonesia is the third largest after oil and coal. Increasing the amount of natural gas needs is directly proportional to the use of natural gas for the energy and industrial raw materials, as well as for domestic purposes. Therefore, there is a need to develop the existing natural gas processing to produce a better quality gas. Natural gas quality is usually determined by the composition of the substances contained in the gas. In general, the content of natural gas is methane (CH₄), ethane (C₂H₆), propane (C₃H₈), iso-butane (iC₄H₁₀) and n-butane (nC₄H₁₀), and non-hydrocarbons such as carbon dioxide (CO₂), nitrogen (N₂) and hydrogen sulfide (H₂S) [2]. The content of impurities such as CO₂ must be lowered as low as possible as it can cause corrosion in gas piping [3], reduce the calorific value of the gas [1], and clog...
the piping system [4]. The CO₂ content in natural gas can vary from place to place and in Indonesia ranging from 5 to 75% depending on the location. As for example, the content of 15% CO₂ in natural gas obtained in the Madura Strait, while the content of up to 75% in natural gas obtained in the Natuna Islands.

The technique has been used so far for the removal of CO₂ from natural gas is a chemical and physical absorption, solid adsorption, cryogenic distillation and membrane separation [5]. The most widely used technology is absorption. The absorption process is conventionally using packed towers and plate columns and it has high value for the mass transfer coefficient. However, this operating unit has disadvantages in its operation such as entrainment, flooding, foaming, small contact area per unit volume and high operating costs.

Therefore, there is a need of alternative technologies which more effective to absorb CO₂ from the natural gas. One of the alternative technologies for the removal of CO₂ from the natural gas is the membrane contactor. This technology is able to overcome the problems that arise in the previous separation technology and this technology also requires only a fairly low energy and the process is much simpler [6]. The separation process using a membrane contactor has several of important factors that affects the performance such as absorbent and feed gas flow rates and the concentration of absorbent [5]. This study will utilize super hydrophobic hollow fiber membrane module as a contactor to absorb CO₂ from its mixture with CH₄ or N₂ using physical absorbent polyethylene glycol (PEG). The physical absorbent has the advantages as it is not corrosive and energy required for the regeneration is lower compared to chemical absorption [7]. The objectives are to see the effect of feed gas flow rate and the number of fibers in the contactor on the mass transfer coefficient, the flux and the absorption efficiency.

2. Materials and Methods
The CO₂ absorption experiment is schematically shown in Figure 1 which has been presented elsewhere [8, 9]. There were three membrane contactors applied in this study consists of 1000, 3000 and 5000 fibers sized 6 and 25 cm in diameter and length, respectively. The contactors were super hydrophobic hollow fiber membrane modules which are provided by PT GDP Filter Bandung. The fibers are polypropylene based sized 525 and 235 μm in outer and inner diameter, respectively. The feed gas is a mixture or CO₂ and CH₄ (36:64) or CO₂ and N₂ (13:87), whilst the absorbent is solution of 5 vol. % PEG in water. The feed gas containing CO₂ was flowed through the shell side of the contactor, whilst the absorbent was pumped through the lumen fiber in the contactor in a counter current arrangement. The inlet and outlet gases to and from the contactor were measured using mass flow meter Sierra Top Trak Instruments, whilst the inlet absorbent to the contactor were measured using liquid flow meter Krohne. Meanwhile, the inlet and outlet gases composition were analyzed using gas chromatography Bruker Scion 436-GC.

The overall mass transfer coefficient, $K_{OVL}$, and CO₂ absorption efficiency, %$R$, were calculated using the following equations [10]:

$$K_{OVL} = \frac{Q_{gin} \ln (c_{in}/c_{out})}{A_m}$$

(1)

$$%R = \frac{c_{in} - c_{out}}{c_{in}} \times 100\%$$

(2)

Meanwhile, CO₂ flux through the membrane contactor, $J$, [9] and acid loading were calculated by:

$$J = (Q_{gin} - Q_{gout}) \times RT / A_m$$

(3)

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

(4)
3. Results and Discussion

Figure 2 shows the CO₂ overall mass transfer coefficients as a function of the feed gas flow rate at different number of fibers in the membrane contactor when absorbent flow rate in the lumen fibers was set at 300 ml.min⁻¹. Figure 2 reveals that the CO₂ overall mass transfer coefficient increases with increasing the feed gas flow rate in the shell side of the contactor due to more CO₂ can be absorbed by absorbent solution flowing in the lumen side of the contactor [11]. The similar trend also shows by the previous study using DEA vol.5% solution as absorbent for CO₂-N₂ (13:87) and CO₂-CH₄ (36:64) system [9] and by Wang et al., [11] using 2M DEA solution as absorbent for CO₂-N₂ system (20:80). In this study the overall mass transfer coefficient increases from 2.6 to 4.3 x 10⁻⁵ cm.s⁻¹ and from 1.3 to 5.8 x 10⁻⁵ cm.s⁻¹ for CO₂-CH₄ and CO₂-N₂ system, respectively, if the gas flow rate increase from 130 to 380 ml.min⁻¹. In the previous study [9] the overall mass transfer coefficient increased from 23 to 32 x 10⁻⁵ cm.s⁻¹ and from 1.39 to 37 x 10⁻⁵ cm.s⁻¹ for CO₂-CH₄ and CO₂-N₂ system, respectively, if the gas flow rate increased from 120 to 340 ml.min⁻¹ using DEA vol.5% solution as absorbent. Meanwhile, Wang et al., [11] reported that the overall mass transfer coefficient increased from 1.6 to 2.5 m.s⁻¹ if the gas velocity increased from 0.03 to 0.09 m.s⁻¹ for CO₂-N₂ system (20:80) using 2M DEA solution as absorbent.

The effect of the number of the fiber in the contactor can also be seen in Figure 2. Figure 2 shows that the mass transfer coefficient decreases with an increase in the number of fiber in the contactor at the same feed gas flow rate. The increase in the number of fiber will increase the surface area for gas-liquid contact, which will enhance the mass transfer coefficient, but will decrease the absorbent velocity in the lumen fiber, which will reduce the overall mass transfer coefficient. The decrease in the overall mass transfer coefficient with increasing the number of fibers in the contactor reveals that the effect of absorbent velocity more dominant than the effect of surface area for gas-liquid contact.
The CO\textsubscript{2} overall mass transfer coefficient, $K_{OVL}$, using 5 vol.% PEG solution as absorbent on the lumen side and a mixture of CO\textsubscript{2}-CH\textsubscript{4} (CH\textsubscript{4}-1000, CH\textsubscript{4}-3000 and CH\textsubscript{4}-5000) or a mixture of CO\textsubscript{2}-N\textsubscript{2} (N\textsubscript{2}-1000, N\textsubscript{2}-3000 and N\textsubscript{2}-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers.

Figure 3 demonstrates the variation of the fluxes as a function of the feed gas flow rate at different number of fibers in the contactor when absorbent flow rate in the lumen fibers was set at 300 ml.min\textsuperscript{-1}. In addition to the mass transfer coefficient, the CO\textsubscript{2} flux increases with increasing the feed gas flow rate in the contactor due to more CO\textsubscript{2} can be absorbed by absorbent solution flowing in the lumen side of the contactor [11]. The similar trend was also shown by previous study [9] and Wang et al. [11]. In this study the CO\textsubscript{2} flux increases from 1.05 to 1.73 mol.m\textsuperscript{-2}.s\textsuperscript{-1} and from 0.49 to 2.18 mol.m\textsuperscript{-2}.s\textsuperscript{-1} for CO\textsubscript{2}-CH\textsubscript{4} and CO\textsubscript{2}-N\textsubscript{2} system, respectively, if the gas flow rate increases from 130 to 380 ml.min\textsuperscript{-1}. In the previous study [9] the CO\textsubscript{2} flux increased from 2.6 to 3.9 x 10\textsuperscript{-6} mmol.cm\textsuperscript{-2}.s\textsuperscript{-1} and from 2.7 to 8.8 x 10\textsuperscript{-6} mmol.cm\textsuperscript{-2}.s\textsuperscript{-1} for CO\textsubscript{2}-CH\textsubscript{4} and CO\textsubscript{2}-N\textsubscript{2} system, respectively, if the gas flow rate increased from 120 to 340 ml.min\textsuperscript{-1} using DEA vol.5% solution as absorbent. Meanwhile, Wang et al., [11] reported that the flux of CO\textsubscript{2} increased from 3 to 8 x 10\textsuperscript{-4} mol.m\textsuperscript{-2}.s\textsuperscript{-1} if the feed gas velocity increased from 0.03 to 0.09 m.s\textsuperscript{-1} for CO\textsubscript{2}-N\textsubscript{2} system (20:80) using 2M DEA solution as absorbent. The CO\textsubscript{2} flux decreases with increasing the number of fibers in the contactor, as illustrates in Figure 3, indicating that the effect of absorbent flow rate more dominant than the effect of the surface area for gas-liquid contact.

The variation of CO\textsubscript{2} absorption efficiency, $R$ (%), as a function of the feed gas flow rate at different number of fibers in the membrane contactor when absorbent flow rate in the lumen fibers was set at 300 ml.min\textsuperscript{-1} is presented in Figure 4. The different results were demonstrated by the two feed gas system. For the CO\textsubscript{2}-N\textsubscript{2} system the CO\textsubscript{2} absorption efficiency increases with the feed gas flow rate. Meanwhile for the CO\textsubscript{2}-CH\textsubscript{4} system the CO\textsubscript{2} absorption efficiency decreases with the feed gas flow rate. The reduction in the CO\textsubscript{2} absorption efficiency was also shown in the previous study [9] and by Yan et al. [5]. In this study, the CO\textsubscript{2} absorption efficiency for CO\textsubscript{2}-CH\textsubscript{4} system decrease from 7.96 to 5.67% if the gas flow rate increases from 183 to 380 ml.min\textsuperscript{-1}. In the previous study [9], the CO\textsubscript{2} absorption efficiency for CO\textsubscript{2}-CH\textsubscript{4} system decrease from 72.7 to 64.0% if the gas flow rate increase from 170 to 340 ml.min\textsuperscript{-1} using DEA vol.5% solution as absorbent. Meanwhile, Yan et al., [5] reported that the CO\textsubscript{2} absorption efficiency decrease from 94 to 65% and 89 to 54% if the gas velocity increase from 0.21 to 0.56 m.s\textsuperscript{-1} using 1M PEG and 1M MEA as absorbents, respectively. The number of fibers in the contactor has the same effect on the CO\textsubscript{2} absorption efficiency for both feed gas system. The CO\textsubscript{2} absorption efficiency increases with the number of fibers in the contactor.
Figure 3 The CO₂ flux, \( J \), using 5 vol.% PEG solution as absorbent on the lumen side and a mixture of CO₂-CH₄ (CH₄-1000, CH₄-3000 and CH₄-5000) or a mixture of CO₂-N₂ (N₂-1000, N₂-3000 and N₂-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers.

Figure 4 The CO₂ absorption efficiency, \( R \) (%), using 5 vol.% PEG solution as absorbent on the lumen side and a mixture of CO₂-CH₄ (CH₄-1000, CH₄-3000 and CH₄-5000) or a mixture of CO₂-N₂ (N₂-1000, N₂-3000 and N₂-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers.

The variation of CO₂ loading as a function of the feed gas flow rate at different number of fibers in the membrane contactor when absorbent flow rate in the lumen fibers was set at 300 ml.min⁻¹ is presented in Figure 5. The CO₂ loading is the ratio between the amounts of CO₂ absorbed to the mole of absorbent flowing in the membrane contactor. As for example in the conventional column, the ideal CO₂ loading using amine solution as absorbent is 0.5 according to the equation below:

\[
2RNH₂ + CO₂ + H₂O \xrightarrow{49 °C} (RNH₂)₂H₂CO₃
\] (5)
The CO$_2$ loading in this study increases with the feed gas flow rate and the number of fibers in the contactor due to more CO$_2$ absorbed by the absorbent as a result of the increase in the feed gas flow rate and the surface area for gas-liquid contact.

![Graph showing CO$_2$ loading vs. Qg (mL/min$^{-1}$)](image)

Figure 5 The CO$_2$ loading using 5 vol.% PEG solution as absorbent on the lumen side and a mixture of CO$_2$-CH$_4$ (CH$_4$-1000, CH$_4$-3000 and CH$_4$-5000) or a mixture of CO$_2$-N$_2$ (N$_2$-1000, N$_2$-3000 and N$_2$-5000) on the shell side of the contactors containing 1000, 3000 and 5000 fibers.

4. Conclusion

The process of gas-liquid contact through the membrane contactor can overcome the disadvantages of the conventional contactor using column or tower such as entrainment, flooding, unloading and foaming. The membrane gas-liquid contactor also requires low energy and the process is much simpler than conventional gas-liquid contactor. This study utilized the super hydrophobic hollow fiber modules as contactors to absorb CO$_2$ using 5 vol.% PEG as absorbent. Feed gas flow rate and the number of fibers in the contactor were the main observed variables to see their effects on the overall mass transfer coefficient, flux, absorption efficiency and the CO$_2$ loading. The experimental results showed that the mass transfer coefficient, flux, absorption efficiency for CO$_2$-N$_2$ system and CO$_2$ loading increased with increasing the feed gas flow rate, but the absorption efficiency for CO$_2$-N$_2$ system decreased. The mass transfer coefficient and the flux, at the same feed gas flow rate, decreased with increasing the number of fibers in the membrane contactor. Meanwhile, the absorption efficiency and the CO$_2$ loading, at the same feed gas flow rate, increased with increasing the number of fiber in the membrane contactor.

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