Transversal Submerged Membrane Contactor for Simultaneous Absorption and Desorption of CO₂ During Natural Gas Sweetening Process

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Abstract. In this work, a unique transversal submerged membrane contactor is used for CO₂ removal. The developed configuration is aimed to provide the possibility to perform simultaneous absorption and desorption of carbon dioxide in a single module. In addition, this configuration is expected to reduce the solvent circulation which is the major obstacle encountered in separated absorption-desorption unit. The module contains a number of polypropylene hollow fiber membranes arranged perpendicularly to the longitudinal axis in which the fibers are submerged into a solvent. Pure water which is placed in the shell of the module is used as the solvent. The effects of operating parameters such as gas flow rate (40-80 ml/min) and operating time on membrane performance are investigated. In addition, the simultaneous absorption-desorption process is compared to the separated ones. Results revealed that CO₂ removal is significantly affected by the gas flow rate. The optimum gas flow rate is 60 ml/min (vacuum pressure of 60.5 kPa) with >80% CO₂ absorption and desorption efficiencies. During a long-term operation, the separation efficiencies are decreased after 200 minutes of operation which may be associated with membrane wetting phenomenon. The simultaneous absorption-desorption process shows a higher separation efficiency rather than the separated process.

1. Introduction

In natural gas sweetening process, CO₂ should be reduced to a certain level to prevent several problems including a freezing in low-temperature chillers, corrosion, reduction of the caloric value, and reduction of pipeline capacity [1]. There are several methods that have been used to remove CO₂ such as solvent-based absorption, solid adsorption, and membrane-based separation. Absorption using amine-based solvent is currently the well-established method. It also has a high selectivity and a high absorption capacity [2,3]. However, conventional absorption column has several disadvantages such as high energy for solvent regeneration, difficulty designing the gas-liquid mass transfer area, and a limited range of gas and liquid flow rates due to operational problems e.g. flooding, loading, channeling, and foaming [4,5]. Membrane technology has been considered as the attractive alternative and has started to replace the conventional technologies in various processes including separation,
chemical reaction, and contacting processes [6–14]. Membrane offers advantages such as relatively lower energy consumption, relatively low operating cost, smaller footprint, easy to scale-up, and easy to operate which make the rapid development and applications of membrane in almost industrial sector [15–20]. In addition, membrane technology also provides a more intensive process and can be combined with several processes into a hybrid process thus the processing steps can be reduced into a single unit [21–25]. Membrane contactor is a hybrid process that combines the advantages of selective absorbent with membrane contactor. The use of membrane as contacting device is aimed to solve the operational problems encountered in conventional absorption column. However, in a separated adsorption and desorption process, circulation of a large volume of solvent is needed for a continuous process which will increase the operational expenditure due to the high cost of solvent. Therefore, development of new configuration is needed to solve this problem.

In this study, a new configuration of transversal submerged membrane contactor is proposed to overcome operational problems of the separated adsorption and desorption process for CO$_2$ removal. A transversal module is used to improve the separation performance of membrane contactor. Submerged membrane configuration is expected to avoid solvent recirculation and reduce the operating cost. Moreover, a simultaneous desorption or solvent regeneration is aimed to improve mass transfer rate during the absorption process.

2. Materials and method
Hollow fiber polypropylene (PP) membrane (supplied by GDP Filter, Indonesia) was used to fabricate membrane module. The PP fiber has 0.35 mm inner diameter, 0.5 mm outer diameter, and 0.2 μm pore size. The total surface area of the membrane was 0.27 m$^2$. Pure CO$_2$ gas (obtained from a local supplier) was used as the feed gas. Pure water was used as solvent (absorbent).

Experimental set up used in this study is shown in Figure 1. The set-up consists of CO$_2$ vessel, membrane module, trap, and vacuum pump (Figure 1 (a)). Pure water is placed in the shell of the module wherein the membranes are submerged into the solvent. The module contains of hollow fiber PP which is arranged perpendicularly to longitudinal axis of the module (Figure 1 (b)). Moreover, the fibers comprise of two layers for feed and permeate gas (Figure 1 (c)). Pure CO$_2$ gas is transferred into PP fiber lumen of the first layer at 40-80 ml/min (Figure 1 (d)). Then, the gas diffuses into the shell side of the module via membrane pores. In the shell side, the gas is absorbed by the solvent. Simultaneously, the absorbed CO$_2$ is desorbed through the adjacent fiber or the second layer by applying vacuum pressure (60.5 kPa) (Figure 1 (d)). In this way, the simultaneous adsorption-desorption process occurs. The amount of CO$_2$ absorbed in the solvent is analyzed by titration method using sodium hydroxide solution (0.1 M). Performance of the membrane contactor is analyzed based on CO$_2$ flux and removal efficiency.

Figure 1. Experimental set-up. (a) Schematic of CO$_2$ removal experiments, (b) transversal membrane contactor, (c) top view of transversal membrane contactor, and (d) absorption-desorption process in submerged transversal membrane contactor.
3. Results and discussion

Performances of membrane contactor in absorption and desorption processes are shown in Figure 2 (a) and (b). It can be observed that CO$_2$ removal is increased when the gas flow rate is increased from 40 to 60 ml/min. The increasing CO$_2$ removal may be associated with a decreasing boundary layer thickness which leads to a better gas diffusion from membrane side to the solvent in the shell side. However, a further increase of the gas flow rate results in a reduction of CO$_2$ removal. This is due to a shorter residence time of gas in the membrane which reduces the transport of CO$_2$ from gas phase to the liquid phase. Meanwhile, CO$_2$ flux increases with the increase of gas flow rate. This may be due to the fact that the amount of CO$_2$ increases as the gas flow rate is increased. In desorption, both CO$_2$ flux and separation decrease along with increasing gas flow rate. The decreasing desorption efficiency may be associated with the increasing amount of CO$_2$ absorbed in the solvent. Since the amount of absorbed CO$_2$ is higher while the vacuum pressure is constant, the amount of desorbed CO$_2$ is almost the same. Consequently, the desorption rate is reduced. In addition, the declining CO$_2$ flux at higher gas flow rate may be attributed to the reduction of the gas film layer on the membrane surface. This will reduce the mass transfer rate during the desorption process.

![Figure 2](image1.png)

**Figure 2.** The effect of gas flow rate on CO$_2$ flux and removal efficiency during (a) absorption and (b) desorption process.

Figure 3 shows the performance of membrane contactor at the various operating time. The performance of the membrane is relatively stable during the first 200 min. The performance of membrane declines both in absorption and desorption processes after 200 min operating time. The declining absorption rate (CO$_2$ removal and flux) may be due to a wetting phenomenon. Wetting gives additional mass transfer resistance which reduces or restricts the diffusion of gas through the membrane. Therefore, the amount of CO$_2$ absorbed and the CO$_2$ flux decrease during this period. The wetting is proved by the result of CO$_2$ concentration analysis in the solvent which is still below the maximum solubility. Since the adsorption and desorption occur simultaneously, the declining absorption rate also leads to decreasing desorption rate. Therefore, during this period, the desorption performance also deteriorates along with declining absorption performance.

![Figure 3](image2.png)

**Figure 3.** Flux and removal efficiency of (a) absorption and (b) desorption process at various operating time.
The performance of simultaneous adsorption-desorption process in the transversal submerged membrane contactor is compared to the separated process. Table 1 shows the comparison of CO₂ removal in separated and simultaneous adsorption-desorption process in a submerged transversal membrane contactor obtained in this study. It can be observed that the simultaneous process provides a better CO₂ absorption and desorption efficiency. Since the solvent is continuously regenerated, the solvent is expected to be in unsaturated condition. Accordingly, the mass transfer rate is better. Therefore, the simultaneous process gives a better separation efficiency rather than the separated one. Furthermore, since the absorbent is stagnant in the membrane module, it gives a sufficient residence time for the absorbent in the module. The longer contact time between the membrane and the absorbent provides the opportunity for the absorbent to achieve a higher absorption capacity. Therefore, a maximum CO₂ removal rate can be achieved. In addition, this also provides the sufficient time for absorbent regeneration process. As the result, both CO₂ absorption and desorption in the simultaneous process are improved. In addition, this novel configuration can eliminate solvent recirculation which is one of the disadvantage of conventional absorption process. Also, another advantage of this configuration is no additional stripping unit is required as encountered in general module [26].

Table 1. Comparison of separated and simultaneous absorption-desorption in transversal submerged membrane contactor.

| Process                      | Operational conditions          | CO₂ removal rate (%) |
|------------------------------|--------------------------------|----------------------|
| Absorption                   | \(Q_\text{gas} = 70 \text{ ml/min}; Q_\text{l} = 109 \text{ ml/min};\) | 46.3                 |
| Desorption                   | \(Q_\text{gas} = 204 \text{ ml/min}; P_\text{vacuum} = 34.5 \text{ kPa};\) | 45.8                 |
| Absorption-desorption        | \(Q_\text{gas} = 60 \text{ ml/min}; P_\text{vacuum} = 60.5 \text{ kPa};\) | Absorption >80%      |
| (submerged membrane)         | \(\)                            | Desorption > 80%     |

4. Conclusion
In this work, a unique transversal submerged membrane contactor is used for CO₂ removal. The developed configuration is aimed to provide the possibility to perform simultaneous absorption and desorption of carbon dioxide in a single module. In addition, this configuration is expected to reduce the solvent circulation which is the major obstacle encountered in separated absorption-desorption unit. Results revealed that CO₂ removal is significantly affected by the gas flow rate. The optimum gas flow rate is 60 ml/min (vacuum pressure of 60.5 kPa) with >80% CO₂ absorption and desorption efficiencies. During a long-term operation, the separation efficiencies are decreased after 200 minutes operation which may be associated with membrane wetting phenomenon. The simultaneous absorption-desorption process shows a higher separation efficiency rather than the separated process.

5. References
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