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Effect of Diethanolamine (DEA) Solvent Flow Rate on the CO_2 Absorption-Desorption Process Using a Hollow Fiber Membrane Contactor

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ABSTRACT: One of the primary objectives in decarbonization is the separation of CO_2 from industrial gas mixtures, particularly in application such as biogas purification and flue gas treatment. A dual-layer crossflow membrane module was utilized under both circulation and batch operating modes with a 30% DEA solution. This study investigates the influence of solvent flow velocity on CO_2 separation performance using a hollow fiber membrane contactor with a 30% DEA solvent. the process was evaluated under two operating modes: batch and solvent circulation. Key variables measured include the solvent flow rate (40–160 mL/min), operating temperature (30–50°C), and sweep gas flow rate (100–300 mL/min). The results indicate that under continuous operation with a solvent flow rate of 160 mL/min, a temperature of 30°C, and a sweep gas flow rate of 100 mL/min, 50.42% of the CO_2 was successfully removed. In contrast, the batch system, under identical conditions achieved only a 27.8% removal rate. The superior performance in circulation mode is attributed to the continuous renewal of the solvent, which sustains a stable concentration gradient and minimizes mass transfer resistance. These findings underscore the potential of membrane-based systems with optimized solvent circulation for efficient and stable CO_2 capture in industrial applications.
Keywords: carbon dioxide; diethanolamine; membrane contactor; solvent circulation

1. Introduction

Natural gas is a major energy resource that plays a significant role to fulfilling global energy demand. It is widely used as a fuel in power plants, various energy generation facilities, and for household needs (Rahmawati et al., 2023). Its strategic role of natural gas as a transitional energy source positions it a key component in addressing the growing global energy demand while reducing carbon emissions. However, natural gas typically contains impurities such as carbon dioxide (CO_2), which can cause pipeline corrosion, increase maintenance costs, and reduce the calorific value of the gas (Santallum et al., 2021; Ng et al., 2024). Therefore, the removal of CO_2 is essential to ensure the safe, efficient, and sustainable use of natural gas.

A widely studied method for CO_2 separation is absorption, a process in which gas is captured by binding it to a liquid absorbent, typically in the presence of a solvent (Nyamiati et al., 2023). Absorption is generally classified into two types: physical absorption and chemical absorption (Sussatrio et al., 2024). Beyond absorption, other CO_2 separation techniques include adsorption and physical separation methods, such as cryogenic separation and membrane technology (Madejski et al., 2022). Within this context, the membrane contactor emerges, the membrane contactor emerges as a promising alternative to conventional absorption methods. It enables the transfer of acid gases into the absorbent solvent through a porous structure that enhances mass transfer efficiency. Compared to alkanolamine-based absorption processes, membrane contactors offer significantly lower energy consumption. Moreover, they provide several distinct advantages: they are lightweight, compact, and modular; offer a high surface-area-to-volume ratio; and require a relatively low initial capital investment (Ibrahim et al., 2018).

In the context CO_2 capture, gas-liquid membrane-based contactor have been applied in a variety of module designs and configurations. Membrane contactors categorized into two operational models: parallel and cross-flow. The configuration of flow in gas membranes plays a critical role in determining the efficiency of the separation process. (Scholes & Shen, 2018). A study has been carried out by (Ng et al., 2024) focused on modelling hollow fiber-based membrane contactors featuring two configurations: crossflow and parallel. The results obtained show that the increasing the percentage of cross-flow to the configuration, the more CO_2 gas is absorbed. In 2017, Chang, et al. simulated a cross-flow membrane system designed to handle the concurrent absorption and desorption processes of CO_2 gas. The results obtained show that the cross-flow membrane configuration offered higher efficiency, with more consistent mass flux in several parts of absorption and stripping.

A study by Chavan et al., (2021) focused on membrane contactors for separating CO_2 from N_2 and H_2 mixtures with a 35/65% volumetric ratio. Using a polypropylene membrane and the pH swing absorption method, the study demonstrated a promising recovery of N_2 and H_2. However, a limitation of this research was the requirement for large tanks, as the absorption and desorption processes do not occur in a single location. Studied conducted by Chang et al., (2017) and Sohaib et al., (2020) explored the simultaneous separation of CO_2 gas through absorption and desorption mechanisms using a hybrid membrane contactor, as well as the use of ionic liquids for concurrent CO_2 capture. Although this method is capable of separating CO_2, its efficiency level remains relatively lower compared to conventional chemical absorption processes.

The simultaneous absorption and desorption of CO_2 using a novel hybrid membrane contactor (HASMC) that combines parallel-type and contiguous-type module configurations was the focus of a study conducted by Pan et al in 2017 Propylene carbonate was used as the physical solvent in this study to assess CO_2 capture performance using both simulation and experimental methods. Although the hybrid configuration demonstrated a higher absorption flux compared to conventional single-mode absorption systems, its overall efficiency remains limited. due to the relatively lower performance in low-concentration CO_2 conditions when compared to traditional chemical absorption methods.

The membrane contacts technique employs a hydrophobic microporous membrane to as a stable interface that separates the gas and liquid phases. This facilitates mass transfer while avoiding direct mixing. In Figure 1, the transfer of CO_2 takes place in three distinct stages. The initial step involves the gas from the main stream diffusing to the outer surface of the membrane, followed by its passage through the membrane's porous structure, and ultimately enters the liquid phase. Next, in Figure 2, desorption takes place, during which the bound CO_2 is released by a sweep gas (typically N_2) through the membrane pores to regenerate the solvent. (Waseem et al., 2025).

The overall mass balance for the simultaneous CO_2 absorption–desorption process can be derived based on the flow diagram presented in Figure 3. This process involves two main phases: the gas phase (feed gas), the liquid phase (solvent), the product gas (sales gas). Assuming steady-state conditions and no mass accumulation within the system, the total mass balance equation can be written as follows.

$$OMB = input - Output \quad (1)$$

$$OMB = Feed + SG_{in} - Sales - L_{out} \quad (2)$$

Next, the mass balance for the CO_2 component, as the target component in this separation process, can be written as follows:

$$In = F \times X_f + L_{in} \times X_{L_{in}} \quad (3)$$

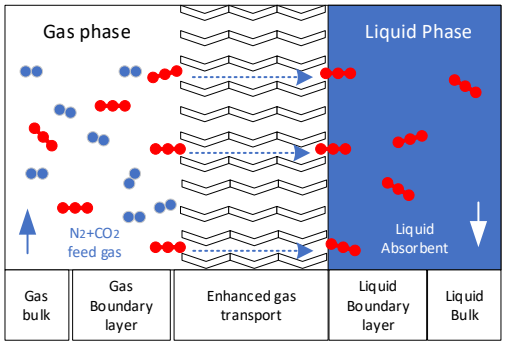

Figure 1. The transport behavior of CO_2 during absorption across a porous membrane system

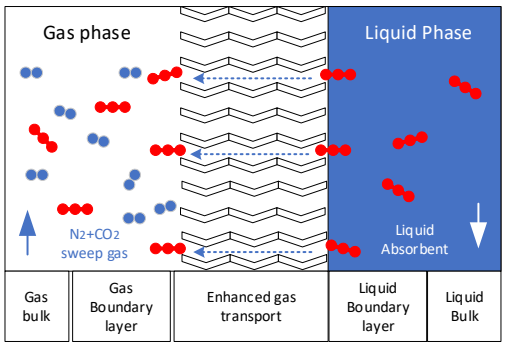

Figure 2. Mechanism of CO_2 gas mass transfer in the desorption process on a porous membrane

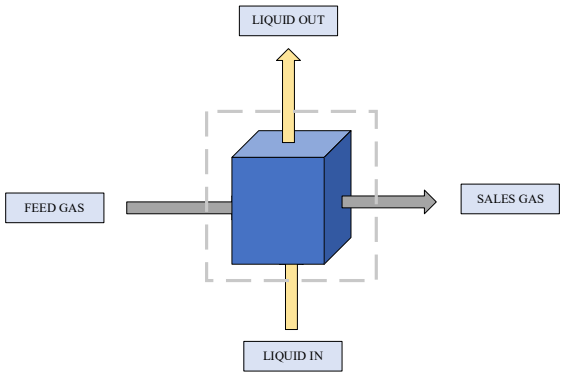

Figure 3. Flow Diagram of the Porous Membrane Unit designed for Concurrent Absorption and Stripping Process of Carbon Dioxide

$$Out = Sales \times X_{sales} - L_{out} \times X_{L_{out}} \quad (4)$$

Assuming the system operates under a equilibrium condition, solvent capacity is not a limiting factor, and there is no accumulation of CO_2 in the solvent. Thus, the mass balance equation becomes:

$$F \times X_f = Sales \times X_{sales} \quad (5)$$

Therefore, the calculation of CO_2 removal efficiency can use the equation:

$$\eta = \frac{CO_{2in} - CO_{2out}}{CO_{2in}} x\ 100\% \quad (6)$$

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To find the absorption flux, an equation can be used that is derived from the energy balance equation in the research equipment system. The absorption flux equation can be seen in the following equation.:

$$(J_{CO_2})_{Abs} = \frac{(Q_{gas\ in} \times C_{gas\ in}) - (Q_{gas\ out} \times C_{gas\ out})}{A_{Abs}} \quad (7)$$

where JCO_2 is the absorption flux (mol/m^{2}s), Qin and Qout are the flow rates of incoming and outgoing CO_2 gas (m3/s), Cin and Cout indicates the amount of carbon dioxide present in the incoming and outgoing gas line (mol/m^3). Meanwhile, to find the value of the desorption flux, the desorption flux equation is used as follows:

$$(J_{CO_2})_{Des} = \frac{(Q_{Sweep\ gas\ out} \times C_{sweep\ gas\ out})}{A_{Des}} \quad (8)$$

Where C sweep gas out is the concentration of sweep gas and Q sweep gas out is the flow rate of sweep gas.

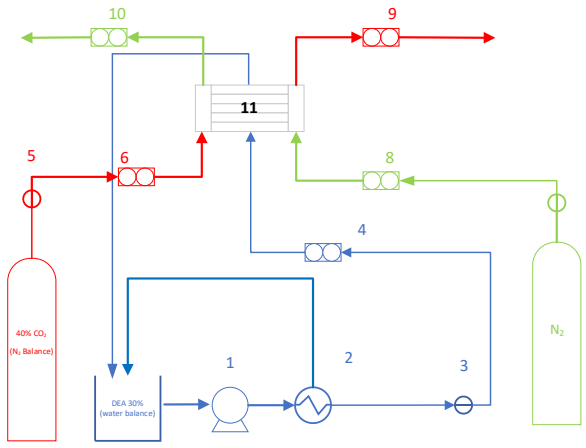

Figure 4. Research Equipment Diagram, 1. Pump, 2. Heater, 3. Pressure gauge, 4. Liquid flowmeter, 5. Pressure gauge, 6. CO_2 gas inflow flowmeter, 7. Pressure gauge, 8. Sweep gas inflow glowmeter, 9. CO_2 gas outflow flowmeter, 10. Sweep gas outflow flowmeter, 11. Membrane contactor.

Although numerous studies have been conducted to examine how flow configuration and module design impact the CO_2 absorption and desorption processes utilizing contactor membranes, research specifically highlighting the performance of simultaneous absorption-desorption processes with the addition of continuous flow and variations in solvent flow rates in a double-crossflow membrane configuration remains limited. Several previous studies have focused more on configuration simulations (Chang et al., 2017) or operational stability testing (Chavan et al., 2021), or the implementation of simultaneous absorption–desorption within a hybrid membrane module using physical solvents (Pan et al., 2017), few have addressed operational factors, such as the influence of solvent circulation systems on process efficiency. The objective of this study is to evaluate the impact of under batch and continuous operating modes in a dual-layer crossflow membrane contactor is the goal of this investigation.

2. Materials and Methods

This research uses hydrophobic polypropylene (PP) membrane material obtained from GDP Filter-Indonesia. CO_2 gas was obtained from PT. Aneka Gas Industri Tbk with a concentration entering the membrane module of 40% (balance N_2). The solvent used for the CO_2 absorption-desorption process is Diethanolamine (DEA) Merck KGaA 99% diluted to 30% by weight. Other materials required for the chittick titration process are 1 N HCl solution, saturated NaCl solution, NaOH solution, and methyl orange. Here are the specifications of the membrane characteristics to be used as presented in Table 1.

Table 1. Characteristics of the membrane constructed from hollow fiber

Parameter	Spesification
Inside diameter	0.35 mm
Outside diameter	0.5 mm
Porous diameter	0.1 µm
Fiber Lenght	10 cm
Porosity	0.65 (unitless)
Absorption Area Surface	94.2 cm^2
Desorption Area Surface	904.32 cm^2

2.1. Experimental set-up

In this study, a simultaneous absorption-desorption process will be executed using a HFMC with a 30% DEA solvent as the solvent.

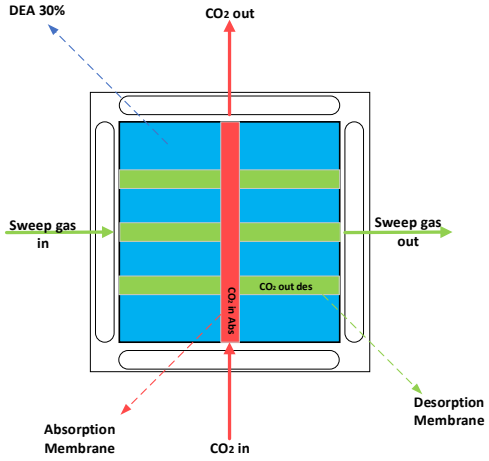

Figure 5. Membrane Arrangement Used in the Research

For the gas absorption process, the incoming gas will contain 40% CO_2 and will flow through the membrane tube

in the first layer, while the solvent in the shell will be continuously circulated. Subsequently, a desorption process will also be carried out simultaneously by drawing the incoming CO_2 gas from the second layer tube of the solvent in the shell, where CO_2 will be carried away by the N_2 sweep gas. The layout of the experimental system is presented Figure 4 and Figure 5.

3. Results and Discussion

3.1. The Effect of Liquid Flow Rate on Overall Efficiency and Absorption-Desorption Flux

Figure 6 illustrates how variations in liquid flow rate impact the system's overall efficiency. Liquid flow rates used are 40, 80, 120, and 160 mL/min. The observation indicates that a higher the liquid flow correlates with enhanced efficiency. The enhancement in overall efficiency correlates with the intensity of the absorption flux.

Flux is defined as the amount of CO_2 that moves per unit time per unit area of the membrane. The greater the solvent flow rate, the greater the absorption flux, resulting in more CO_2 being absorbed in a given time, thereby reducing the CO_2 in the exit gas. This directly increases the efficiency of the system because more CO_2 from the feed gas is successfully removed. This mechanism aligns with established theory, where the Sherwood number (Sh) representing the ratio of convective to diffusive mass transfer increases with Reynolds number (Re) under laminar flow, indicating improved mass transfer efficiency at higher flow rates. This result is consistent with the research of several researchers such as (Lim et al., 2024) and (Li et al., 2021) reported CO_2 removal efficiencies of around 40–60% at lower liquid flow rates using 2.5 M MEA in a PTFE membrane system.

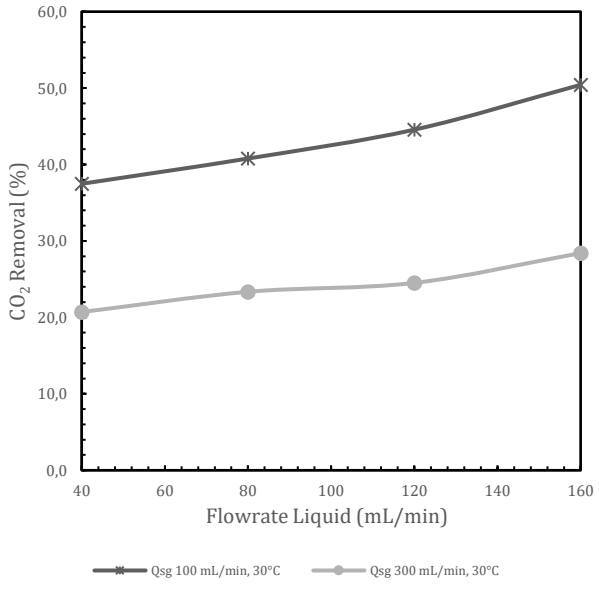

Figure 6. Effect of Liquid Flow Rate on Overall Efficiency

In Figure 7 (a), the data indicates that an increased solvent flow rate significantly enhances the absorption rate. Maximum absorption observed at a flow rate of 160 mL/min. In the absorption process, the feed gas containing a lot of CO_2 passes through the membrane module tube, then diffuses into the solvent through the membrane poresThe transport of CO_2 occurs in three phases: (1) Diffusion of the gaseous component from the bulk phase to the exterior layer of the membrane, (2) penetration through the porous membrane structure up to gas-liquid boundary, and (3) solubility of carbon dioxide into the absorbing liquid. The faster solvent flow rate, the more turbulence in the liquid increases, making the boundary layer thinner, CO_2 gas can transfer from the gaseous stream into the solvent stream thanks to this process.

Table 2. Characteristics of the membrane constructed from hollow fiber

Study	Membrane Type	Solvent	Flow Rate (mL/min)	Absorption flux x 10^3 (mmol/m^{2}s)
Lim et al. (2024)	Polypropilene	MEA (2.5 M)	25–100	0,72
This study	Polypropilene (dual-layer)	DEA (30%)	40–160	0;15

Table 2 compares the absorption flux performance between this study and the work by (Lim et al., 2024). While Lim et al. achieved a maximum absorption flux of 0.72 x 10^3 mmol/m²·s using a polypropylene membrane and 2.5 M MEA at flow rates of 25–100 mL/min, this study achieved a flux of 0.15 x 10^3 mmol/m²·s using a dual-layer polypropylene membrane and 30% DEA at higher flow rates of 40–160 mL/min. The lower flux observed in this study is attributed to the use of DEA, a weaker base than MEA, which typically shows slower reaction kinetics in CO_2 absorption. However, DEA offers advantages in thermal stability and lower degradation, making it more suitable for long-term and cyclic operation. Furthermore, the use of a dual-layer hollow fiber configuration contributes to maintaining a stable concentration gradient, enhancing process stability even under continuous flow (Li et al., 2021).

Figure 7 (b) illustrates that the desorption flux increases with rising solvent flow rates with the maximum flux observed at a flow rate of 160 mL/min. In the CO_2 desorption process, one of the factors that affects the desorption flux is the solvent flow rate. When this layer is thick, the transfer of trapped CO_2 into the gas phase is hindered. However, when the solvent flow rate increases, this layer becomes thinner, reducing the transfer resistance and resulting in CO_2 being more easily released and diffusing into the gas phase. These findings are consistent with the results reported by (Mohammed et al., 2021) and (Khaisri et al., 2011) who observed desorption flux values ranging from 2.0 to 7.0 × 10^{-1} mmol/m²·s when using MEA as the solvent at liquid velocities up to 3.5 cm/s

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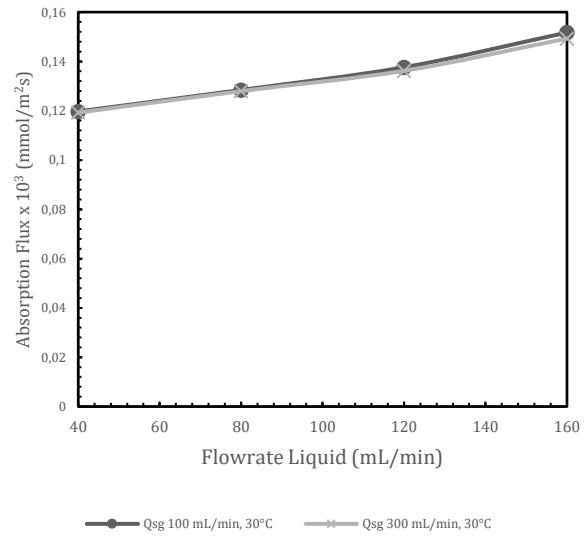
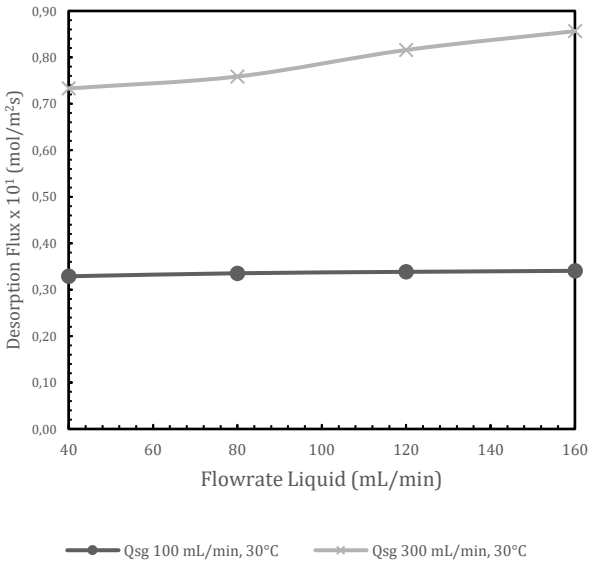


(a)



(b)

Figure 7. Effect of Liquid Flow Rate on the Flux of (a) Absorption and (b) Desorption

3.2. Overall efficiency, absorption and desorption flux in batch operation

Figure 8 shows the overall efficiency values in the batch operation system with variations in the gas sweep flow rate (Qsg) and temperature. From the graph, it is evident that the highest efficiency is achieved at a Qsg condition of 100 mL/min and a temperature of 30°C. Conversely, the lowest efficiency occurs at a Qsg condition of 300 mL/min and a temperature of 50°C. The decreased contact time between the gas and the solvent at higher gas sweep rates limits the effectiveness of CO_2 mass transfer to the solvent and a drop in efficiency. Additionally, the increase in temperature tends to decrease the solubility of CO_2 in the solvent negatively affects the solvent's CO_2 absorption capacity to capture CO_2 even though the reaction rate increases. These findings are consistent with results reported by (Aboshatta & Magueijo, 2021).

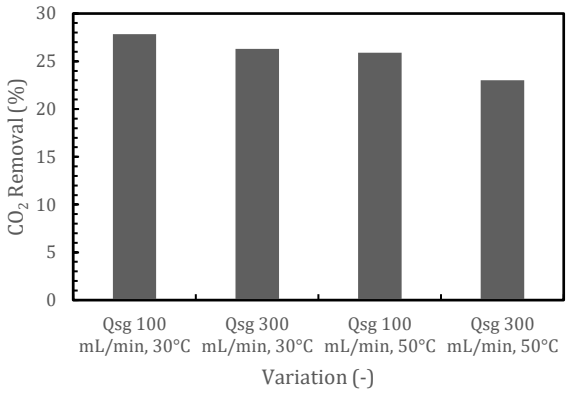
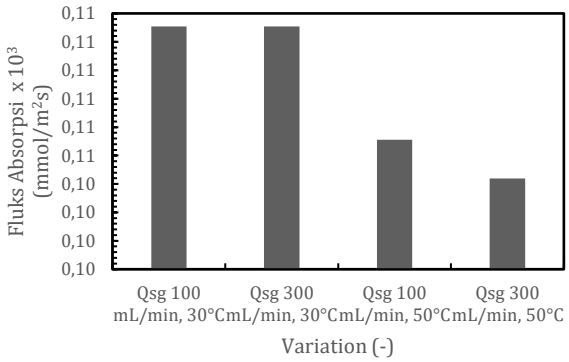
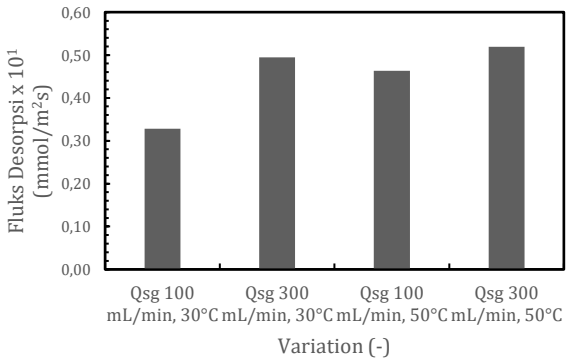


Figure 8. Overall Efficiency in Batch Operations

Figure 9 (a) shows the absorption flux values in a batch operation system with variations in sweep gas flow rate (Qsg) and temperature. As shown in the figure the highest flux is achieved at Qsg 100 mL/min and a temperature of 30°C.



(a)



(b)

Figure 9. Flux (a) Absorption and (b) Desorption Batch Operation (mmol/m^{2}s) with various sweep gas flow rate

Conversely, the lowest flux occurs at Qsg 300 mL/min and a temperature of 50°C as the absorption process performs optimally at low temperatures. Meanwhile, Figure 9 (b) shows that the desorption flux at 30 °C is notably lower compared to 50°C because desorption occurs optimally at high temperatures. These results are consistent with the research by Aboshatta & Magueijo (2021), which demonstrated that performance in membrane contactor systems.

3.3. Comparison of overall efficiency, absorption and desorption flux in circulation and batch operations

Figure 10 presents a comparison of overall CO_2 separation efficiency in the simultaneous absorption-desorption process between batch operation and circulation system with variations in solvent flow rate (QL) from 40 to 160 mL/min. It is evident that the separation efficiency in the batch system exhibits the lowest separation efficiency compared to the circulation system. This is due to the lack of solvent renewal in the batch system, which leads to the gradual saturation of the solvent surface with CO_2 over time.

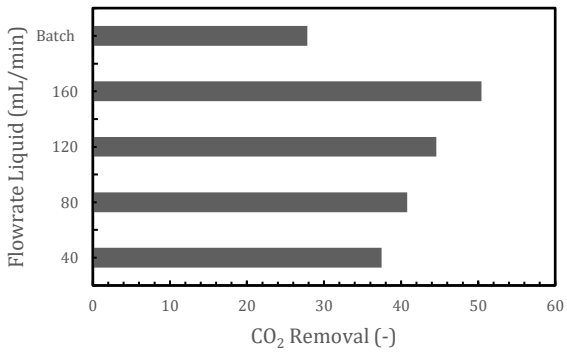


Figure 10. Comparison of Overall Efficiency in Batch and Circulation Operations

The primary force behind mass transfer, the concentration gradient between the gas and liquid phases, is reduced because of this state. In contrast, in a circulation system, the solvent is continuously renewed, keeping the gas-liquid concentration gradient constant. This contributes to the increased separation efficiency along with the increased solvent flow rate. The higher the circulation rate, the faster the saturated solvent is replaced by fresh solvent, which significantly increases the separation efficiency. This trend is clearly visible from the graph, where efficiency gradually increases as the flow rate rises from 40 mL/min to reach its peak at 160 mL/min. These results are consistent with the research of several researchers such as (Lim et al., 2024) and (Li et al., 2021) reported that higher solvent flow rates enhance CO_2 absorption efficiency in CO_2 absorption efficiency, with a flow rate of 25 to 100 mL/min increasing the efficiency from 50% to 90%.

Figure 11 (a) shows that the CO_2 absorption flux in the batch system is lower compared to the circulation system with various solvent flow rates (40 to 160 mL/min). The flux values in the batch system remain low, indicating limited mass transfer during the process. A similar trend is observed in Figure 11(b). The CO_2 desorption flux in the batch system is lower compared to the circulation system at various solvent flow rates. This reduced performance is primarily attributed to the lack of continuous solvent flow which leads to the formation of a thick liquid boundary layer near the membrane surface. This layer becomes the main barrier to CO_2 diffusion between the gas and liquid phases because there is no renewal of the solvent on the membrane surface. As a result, the concentration gradient decreases over time, and the mass transfer coefficient also becomes low.

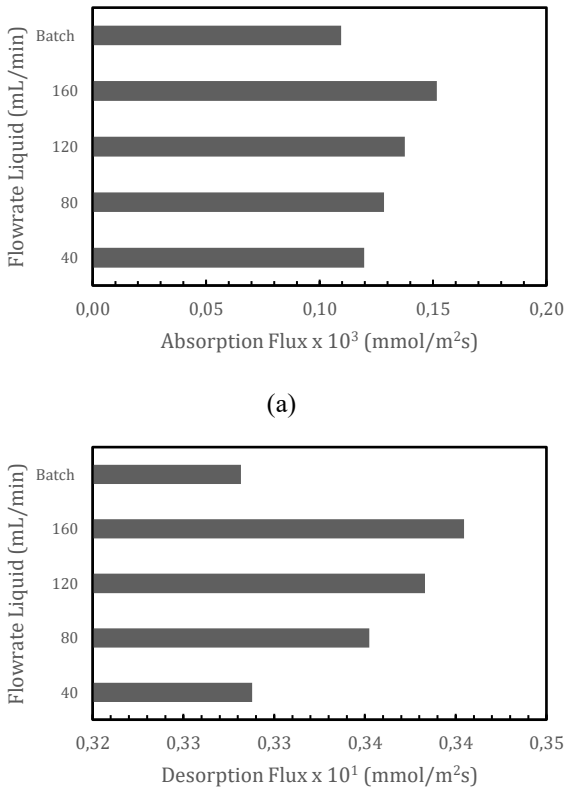


Figure 11. Comparison of Absorption (a) and Desorption (b) Flux in Batch and Circulation Operations

On the other hand, in a circulation system, the solvent is continuously circulated, resulting in the thinning of the liquid boundary layer. This condition enhances mass transfer by reducing diffusion resistance and maintaining a consistent concentration gradient between the liquid and gas phases. As the flow rate increases from 40 to 160 mL/min, the absorption and desorption fluxes significantly increase, indicating that mass transfer occurs more efficiently. This phenomenon confirms that operations with circulating solvents are superior in maintaining the performance of simultaneous CO_2 absorption and desorption processes, particularly in preventing saturation on the solvent surface and enhancing mass transfer capacity. These findings are consistent with the investigations conducted by (Lim et al., 2024) and (Li et al., 2021) regarding CO_2 absorption, as well as (Mohammed et al., 2021) who used MEA solvent for desorption with a membrane contactor.

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4. Conclusions

This research shows that raising the membrane's solvent flow rate significantly enhances separation efficiency and CO_2 flux in the simultaneous absorption–desorption process. The circulation mode of operation exhibited superior performance compared to the batch mode, as continuous solvent renewal helped maintain a stable concentration gradient and reduce mass transfer resistance through boundary layer thinning. Additionally, temperature increase was found to promote the desorption process. Rather than focusing solely on numerical results, these findings demonstrate the potential of membrane-based CO_2 capture systems with optimized solvent circulation for industrial applications, particularly in processes that require operational stability and energy efficiency. This study also lays the groundwork for future research, which could include investigating alternative amine solvents, or incorporating absorption-enhancing elements such as amino acid to further improve performance.

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Credit authorship contribution statement
Raditya Yudhi Pamungkas: Writing: review and editing, writing: initial draft, conceptualization, formal analysis, resources, visualization, validation, and investigation**. Yeni Rahmawati:** Formal analysis, resources, writing, review, and editing; conceptualization; validation**. Siti Nurkhamidah:** Visualization, Project Management, Resources, and Validation**. Fadlilatul Taufany:** Ideas, Materials, Obtaining Funds, and Oversight**. Ali Altway:** Resources, Conceptualization, and Validation**. Susianto:** Data curation, validation, supervision, and methodology.

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