Effect of Temperature and Gas Flow Rate on CO₂ Capture

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INTRODUCTION

Because of the increasing industrialization and the need for energy in the world, our dependence on fossil fuels has increased, which causes an increase in carbon dioxide concentration in the atmosphere. According to NASA, over the past 171 years, the global average amount of carbon dioxide is raised with human activities by 48% above pre-industrial levels found in 1850 and hit a record high in 2021 as 416 parts per million (NASA, 2021). Carbon dioxide (CO₂) is an important greenhouse gas that causes global warming effects (NASA, 2021). To reduce global warming, reduction of CO₂ emissions is critical and considering this energy needs supplied from fossil fuels, CCS (carbon capture storage) is the best way to maintain CO₂ level under control.

Various methods have been studied to reduce post-combustion CO₂ emissions, such as chemical absorption (Heydarifard et al., 2020; Pang et. al., 2021), physical absorption (Elhambaksh et al., 2020), membrane separation (Widakdo et al., 2021; Yu et al., 2021), adsorption (Wu et al., 2021), cryogenic separation (Bi and Ju, 2021), algal system (Judd et al., 2015). In these processes, chemical absorption is extensively used for the removal of CO₂ from flue gas by chemical reaction with a solvent (Chen et al., 2008). The drawbacks of CO₂ adsorption method include adsorption mechanism only occurring on the material surface, and that regeneration would require a stoppage to the adsorption process. Cryogenic CO₂ separation also requires significant amounts of energy for the cooling of CO₂ down to its liquefaction temperature at 78.5 °C. CO₂ capture with a membrane is an interesting field of study but it still faces challenges in the form of the unavoidable tradeoff between permeability and selectivity, as well as poor resistance towards acid gases (Wibowo et al., 2021).

Absorption-based technology is the most widely used worldwide, accounting for 64% of total CO₂ capture capacity in these facilities and is used in 72% of the facilities. Amine-based solvent alone is used in 69% of the facilities, accounting for 55% of total CO₂ capture capacity. The popularity of amine-based absorbent, is most due to its high absorption efficiency, low energy consumption, high economic value, and simple operation (Peng et al., 2012; Wibowo et al., 2021). In the field of CO₂ absorption, innovations can be made on the searching solvents and scrubber types. Several scrubbers are used to capture CO₂, such as packed bed column, sieve tray column and bubble column. Bubble columns have several advantages, such as high mass and heat transfer rates, simple operation without any moving parts and the flexibility to adjust various residence time requirements.

Various solvents are used in the bubble column for CO₂ absorption based on published data. Heydarifard et al. focus on CO₂ reactive absorption using Piperazine aqueous solutions in a stirrer bubble column (Heydarifard et al., 2018). Bai and Yeh (1997) studied the CO₂ capture by ammonia in bubble column reactor. In a stirred bubble column, Pashaei et al. (2017) investigated the solubility and chemical absorption rate of
carbon dioxide into aqueous solutions of diethanolamine. Monoethanolamine (MEA) is frequently used to evaluate experimental setups and as a reference. Aronu et al. (2011) and Maneenir et al. (2009) obtained 76 g CO₂/kg MEA and 80 g CO₂/kg MEA absorption capacities using bubble columns at 20% MEA and 40°C, respectively. Aronu et al. (2011) obtained 0.516 mol CO₂/mol MEA absorption capacity at 2.5 M (%15) MEA, 40°C temperature and 4.84 kpa partial pressure of CO₂.

In this study, the absorption capacity of MEA solution was investigated in a bubble column reactor. The main objective of this study is to determine the effect of temperature and gas flow rate on absorption capacity given as g CO₂/kg solvent and mol CO₂/mol MEA.

**REACTION MECHANISM**

MEA is an important solvent in the CO₂ removal process because it reacts quickly with carbon dioxide due to its primary amine characteristics (Maceiras et al., 2008).

Different reaction mechanisms are proposed based on the number of amine functionality. Two different mechanism named zwitterion and termolecular mechanism is proposed to describe reaction mechanisms between CO₂ and primary amine (RNH₂) solutions. Because of the reaction between CO₂ and MEA solution, the carbamate (RNHCOO⁻) is formed in two steps according to the zwitterion mechanism, in one step according to the termolecular mechanism.

In the zwitterion mechanism, firstly the zwitterion (RNH₂²⁺COO⁻) is formed (Eq. 1) as an intermediate and then it is deprotonated (Eq. 2) to form carbamate (RNHCOO⁻). In these reactions, B could be CO₂⁻, HCOO⁻, amine, H₂O, or OH⁻ (Ramezani et al., 2021).

\[
\begin{align*}
    \text{CO}_2^+ + \text{RNH}_2^+ &\rightarrow \text{RNH}_2\text{COO}^- \quad (1) \\
    \text{RNH}_2\text{COO}^- + \text{B} &\rightarrow \text{RNHCOO}^- + \text{BH}^- \quad (2)
\end{align*}
\]

According to the termolecular mechanism, CO₂ react with MEA, as follow (Eq. 3):

\[
\text{CO}_2 + \text{RNH}_2^- + \text{B} \rightarrow \text{RNHCOO}^- + \text{BH}^- \quad (3)
\]

Additionally, CO₂ reacts with H₂O as given in Eq. (4-6). But overall contribution of these reactions can be negligible in the presence of MEA (Ramezani et al., 2021):

\[
\begin{align*}
    \text{H}_2\text{O} + \text{CO}_2 &\rightarrow \text{H}^+ + \text{HCOO}^- \quad (4) \\
    \text{H}_2\text{O} &\rightarrow \text{H}^+ + \text{OH}^- \quad (5) \\
    \text{HCOO}^- &\rightarrow \text{H}^+ + \text{CO}_3^{2-} \quad (6)
\end{align*}
\]

**MATERIALS AND METHOD**

**Calculation of Absorption Capacity**

The absorption capacity is the maximum amount of pollutant absorbed per amount of solvent. The absorption capacity of continuous system can be calculated using the effluent CO₂ concentration vs time graph. The area in Figure 1 between the curves representing the inlet and outlet concentrations of CO₂ is used to determine the amount of total absorbed CO₂. The upper line is actually the CO₂ concentration in the inlet, which was kept constant during a single run. At the outlet, concentration changes alongside the saturation of the MEA absorbent solution. The difference between these two values shows the absorbed amount. The input flow rate of CO₂ can be calculated using the total flow rate and inlet CO₂ concentration. The outlet flow rate of CO₂ can be calculated using a fixed flow rate of N₂, which is an inert compound and effluent CO₂ concentration. Eq. (7) was used to calculate the CO₂ outlet flow rate:

\[
Q_{CO_2\text{out}} = Q_{total\text{in}} \times \frac{y_{CO_2\text{out}}}{y_{CO_2\text{in}}} \quad (7)
\]

In this equation, \( Q_{CO_2\text{out}} \) is the outlet flow rate (l/min); \( Q_{total\text{in}} \) is the total flow rate (l/min); \( y_{CO_2\text{in}} \) is mol fraction in the gas inlet; \( y_{CO_2\text{out}} \) is CO₂ mole fraction in the gas outlet; \( y_{CO_2\text{out}} \) is N₂ mole fraction in the gas outlet.

The volumetric flow rates are converted to molar mass flow rate using conversion factors and assuming an ideal gas equation of state where each mole of gas occupies 22.4 liters at standard temperature (273 K) and pressure (1 atm). Then, the effluent concentration (ppm)–time graph can be replotted for mass flow rate-time. The rate of absorbed CO₂ at each reading interval is then calculated using Eq. (8):

\[
R_{CO_2} = \frac{M_{CO_2\text{in}} - M_{CO_2\text{out}}}{t_2 - t_1} \quad (8)
\]

where, \( R_{CO_2} \) is the rate of absorbed CO₂ ( l/min); \( M_{CO_2\text{in}} \) is the mass of absorbed CO₂ in the gas inlet, and \( M_{CO_2\text{out}} \) is the mass of absorbed CO₂ in the gas outlet.

The amount of absorbed CO₂ for each time interval was calculated using Eq. (9):

\[
M_{CO_2\text{ab}} = R_{CO_2} \times (t_2 - t_1) \quad (9)
\]

where, \( M_{CO_2\text{ab}} \) is the mass of absorbed CO₂ (mol CO₂), \( R_{CO_2} \) is the rate of absorbed CO₂ (l/min), and \( t \) is the time (0-250 min).

The absorption capacity of the absorbent was calculated using Eq. (10):

\[
\text{Ab. Cap} = \frac{\sum M_{CO_2\text{ab}}}{M_{MEA}} \quad (10)
\]

where \( n \) is the number of time intervals (250 min), \( M_{CO_2} \) is the mass of absorbed CO₂ (mol CO₂), and \( M_{MEA} \) is the mass of MEA (mol MEA) in the solution. The calculations can be done using MS Excel.
Chemicals

MEA was supplied by Sigma (Germany). N₂ (>99.9 %, 200 bar) and CO₂ (>99.95%, 150 bar) gas cylinders were purchased from Oksan gas, Turkey. In all experiments, deionized water was used supplied from the Thermo Scientific (Germany) unit with ultrapure filters.

Experimental Set-up

In this study, the bubble column absorption column shown in Figure 2 was used. The column has a height of 1.00 m and diameter of 5.0 cm. The process was performed in semi batch mode where liquid did not flow (batch) and the gas phase was feeding continuously into the solution. Column surrounded with heat jacket that keeps the solution temperature constant in the column. In each experiment, the temperature of the heat jacket was adjusted to the desired temperature 25 min before the run. However, the MEA solution was heated to the desired temperature on the hot plate and pour into the reactor immediately. A 1L MEA solution is used in each experiment. The gas mixture that consists of CO₂ and N₂ was prepared using two separate mass flow controllers (ALICAT Scientific Mass Flow Controller, Range:0-10L/min, accuracy; %0.2 of full-scale). The gas combination (nitrogen and carbon dioxide) is so dry, a continuous flow of gas through the column would cause the solution to evaporate significantly. This is a negative consequence since the solution’s concentration and absorption capacity will change. As a result, before introducing gas to the absorption column, a humidifier must be used to saturate the incoming gas stream with humidity. The initial CO₂ concentration was adjusted to 50,000 ppm using mass flow controllers. At the beginning of the experiment gas mixture was fed to the CO₂ analyzer to ensure its concentration and then fed to the column filled with MEA solution. The gas mixture was bubbled using diffuser in the column and the carbon dioxide concentration of the effluent gas was monitored using an inline CO₂ gas analyzer (Vernier, USA). The absorption process continued until there was no further absorption. This was confirmed by the concentration/time profile as shown in Figure 1.

RESULTS AND DISCUSSION

The Effect of Temperature on Absorption Capacity

CO₂ absorption capacity is an essential feature for the successful CO₂ removal in a bubble column. The absorption capacity can be expressed in two units as mol of CO₂ absorbed per mol of amine solution and as g CO₂ absorbed per kg of amine solution.

The effect of temperature on the absorption capacity is shown in Figure 3 and Figure 4. The absorption capacity of 66.27 g CO₂/kg solvent and 0.456 mol CO₂/mol MEA decreases to 45.67 g CO₂/kg solvent and 0.32 mol CO₂/mol MEA with increasing temperature from 25 to 45 °C when using %20 MEA solution. This was due to the thermodynamics of the exothermic CO₂ absorption system that could cause reversible reactions when the temperature was too high. The increase in temperature could also increase the CO₂ vapor pressure over the solution that leads to decrease in the physical solubility of CO₂ in the solvent (Tan et al., 2012).

The absorption capacity of 74.71 g CO₂/kg MEA was obtained at 5 L/min gas flow rate, 25°C temperature and 20% solvent concentration. Similarly, Maneerin et al. (2009) obtained 80g CO₂/kg MEA absorption capacity at 20% MEA and 40°C. Aronu et al. (2011) obtained 76 g CO₂/kg MEA absorption capacity at 5.0 M (%20) MEA, 40°C temperature and 15kpa CO₂ with using bubble column.
**The Effect of Gas Flow Rate on Absorption Capacity**

The gas flow rate determines the detention time of the gas component and affects the mixing regime of the solution. The effect of gas flow rate on the absorption capacity is shown in Figure 5 and Figure 6. The amount of CO₂ captured per solvent amount increases with increasing gas flow rate. The absorption capacity of 66.27 g CO₂/kg solvent and 0.456 mol CO₂/mol MEA was increased to 74.71 g CO₂/kg solvent and 0.51 mol CO₂/mol MEA when the gas flow rate increased from 2.5 L/min to 5 L/min. Conway et al. (2015) obtained 0.15 mol CO₂/mol MEA absorption capacity at 4 M (%25) MEA and 3 L/min gas flow rate with using wetted-wall column contactor. Aronu et al. (2011) obtained 0.516 mol CO₂/mol MEA absorption capacity at 2.5 M (%15) MEA, 40°C temperature and 4.84 kpa partial pressure of CO₂ with using bubble column.

With the increasing gas flow rate, detention time of the gas into the solution was decreased. However, a higher flow rate creates turbulent conditions in the column, which is favorable for mass transfer. With increasing gas flow rates, the driving force between CO₂ and water is strengthened and more CO₂ molecules transfer from the bulk gas phase to the gas-liquid boundary layer, causing an increase in mass-transfer coefficient.

**CONCLUSIONS**

In this study, the effect of temperature and gas flow rate on the absorption capacity (g CO₂/kg solvent and mol CO₂/mol MEA) determined using bubble column reactor. Experiments were performed at semi-batch mode and 20% MEA solution was used as a solvent. Because of the study, the following findings were obtained:

1. The absorption capacity increases with the decrease on the temperature. The absorption capacity of 45.67 g CO₂/kg solvent and 0.32 mol CO₂/mol MEA increased to 66.27 g CO₂/kg solvent and 0.456 mol CO₂/mol MEA when the temperature decreases from 45°C to 25°C.

2. Gas flow rate also effect the absorption capacity positively. The capacity increases from 66.27 g CO₂/kg solvent and 0.456 mol CO₂/mol MEA to 74.71 g CO₂/kg solvent and 0.51 mol CO₂/mol MEA when the gas flow rate increases from 2.5 L/min to 5 L/min.

3. Because of the study, absorption capacities of 74.71 g CO₂/kg MEA and 0.51 mol CO₂/mol MEA were obtained at the conditions of 5 L/min gas flow rate, 25°C and 20% MEA concentration. This value is comparable to the values obtained in the literature.

It can be concluded that capture of CO₂ into the MEA solution at bubble column can be successfully achieved with the high absorption capacity. Development of a solvent for a high absorption capacity is one of the most crucial issues for post-combustion capture. Mixed amine solutions can be used to achieve high absorption capacity.

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**Availability of data and materials:** All data generated or analyzed during this study are available for sharing when appropriate request is directed to corresponding author.

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