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Research Article

Effect of Temperature and Gas Flow Rate on CO_2 Capture

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ABSTRACT

In this study, the removal of carbon dioxide (CO_2) that has a huge contribution to global warming from gas emissions was performed using absorption method. Effect of operational parameters such as temperature and gas flow rate on the absorption capacity (g CO_2/kg solvent and mol CO_2/mol MEA) was investigated in a bubble column reactor with a semi-batch operation. The monoethanolamine (MEA) was used as a solvent and absorption capacity was determined at different gas flow rates (2.5 and 5 L/min) and temperatures (25-35-45 °C). Because of the study, absorption capacities of 74.71 g CO_2 / kg MEA and 0.51 mol CO_2/mol MEA were obtained at 5 L / min gas flow rate, 25°C and 20% solvent concentration. The absorption capacity increases as the temperature decreases and as the flow rate increases. Because of the study, it can be concluded that the capture of CO_2 into the MEA solution at bubble column can be successfully achieved with high absorption capacity.

Keywords: MEA, absorption capacity, bubble column, CO_2 fixation

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 (CO_2) is an important greenhouse gas that causes global warming effects (NASA, 2021). To reduce global warming, reduction of CO_2 emissions is critical and considering this energy needs supplied from fossil fuels, CCS (carbon capture storage) is the best way to maintain CO_2 level under control.

Various methods have been studied to reduce post-combustion CO_2 emissions, such as chemical absorption (Heydarifard et al., 2020; Pang et. al., 2021), physical absorption (Elhambakhsh 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 CO_2 from flue gas by chemical reaction with a solvent (Chen et al., 2008). The drawbacks of CO_2 adsorption method include adsorption mechanism only occurring on the material surface, and that regeneration would require a stoppage to the adsorption process. Cryogenic CO_2 separation also requires significant amounts of energy for the cooling of CO_2 down to its liquefaction temperature at 78.5 °C. CO_2 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 CO_2 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 CO_2 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 CO_2 absorption, innovations can be made on the searching solvents and scrubber types. Several scrubbers are used to capture CO_2, 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 CO_2 absorption based on published data. Heydarifard et al. focus on CO_2 reactive absorption using Piperazine aqueous solutions in a stirrer bubble column (Heydarifard et al., 2018). Bai and Yeh (1997) studied the CO_2 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 Maneeintr et al. (2009) obtained 76 g CO_2/kg MEA and 80 g CO_2/kg MEA absorption capacities using bubble columns at 20% MEA and 40°C, respectively. Aronu et al. (2011) obtained 0.516 mol CO_2/mol MEA absorption capacity at 2.5 M (%15) MEA, 40°C temperature and 4.84 kpa partial pressure of CO_2.

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 CO_2/kg solvent and mol CO_2/mol MEA.

REACTION MECHANISM

MEA is an important solvent in the CO_2 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 CO_2 and primary amine (RNH_2) solutions. Because of the reaction between CO_2 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 ion ($RNH_2^+COO^-$) is formed (**Eq. 1**) as an intermediate and then it is deprotonated (**Eq. 2**) to form carbamate ($RNHCOO^-$). In these reactions, B could be CO_3^{2-}, HCO_3^-, amine, H_2O, or OH^- (Ramezani et al., 2021).

$$CO_2 + RNH_2 \leftrightarrow RNH_2^+COO^- \quad (1)$$

$$RNH_2^+COO^- + B \leftrightarrow RNHCOO^- + BH^+ \quad (2)$$

According to the termolecular mechanism, CO_2 react with MEA, as follow (**Eq. 3**):

$$CO_2 + RNH_2 + B \leftrightarrow RNHCOO^- + BH^+ \quad (3)$$

Additionally, CO_2 reacts with H_2O as given in **Eq. (4-6)**. But overall contribution of these reactions can be negligible in the presence of MEA (Ramezani et al., 2021):

$$H_2O + CO_2 \leftrightarrow H^+ + HCO_3^- \quad (4)$$

$$H_2O \leftrightarrow H^+ + OH^- \quad (5)$$

$$HCO_3^- \leftrightarrow H^+ + CO_3^{2-} \quad (6)$$

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 CO_2 concentration vs time graph. The area in **Figure 1** between the curves representing the inlet and outlet concentrations of CO_2 is used to determine the amount of total absorbed CO_2. The upper line is actually the CO_2 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 CO_2 can be calculated using the total flow rate and inlet CO_2 concentration. The outlet flow rate of CO_2 can be calculated using a fixed flow rate of N_2, which is an inert compound and effluent CO_2 concentration. **Eq. (7)** was used to calculate the CO_2 outlet flow rate:

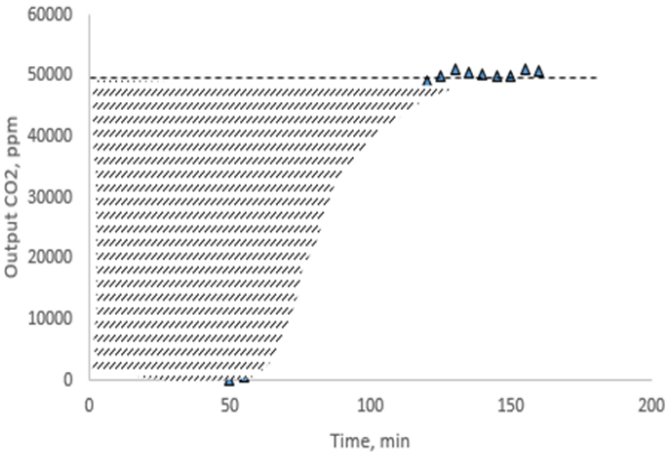

Figure 1. A sample of the CO_2 concentration profile at the output

$$Q_{CO_{2_{out}}} = Q_{total_{in}} \times y_{N_{2_{in}}} \left(\frac{y_{CO_{2_{out}}}}{y_{N_{2_{out}}}}\right) \quad (7)$$

In this equation, $Q_{CO_{2_{out}}}$ is the outlet flow rate (l/min); $Q_{total_{in}}$ is the total flow rate (l/min); $y_{N_{2_{in}}}$ is N_2 mole fraction in the gas inlet; $y_{CO_{2_{out}}}$ is CO_2 mole fraction in the gas outlet; $y_{N_{2_{out}}}$ is N_2 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 CO_2 at each reading interval is then calculated using **Eq. (8)**:

$$R_{CO_2} = \dot{M}_{CO_{2_{in}}} - \dot{M}_{CO_{2_{out}}} \quad (8)$$

where, R_{CO_2} is the rate of absorbed CO_2 (l/min), $\dot{M}_{CO_{2_{in}}}$ is the mass of absorbed CO_2 in the gas inlet, and $\dot{M}_{CO_{2_{out}}}$ is the mass of absorbed CO_2 in the gas outlet.

The amount of absorbed CO_2 for each time interval was calculated using **Eq. (9)**:

$$M_{CO_{2_{ab}}} = R_{CO_2} \times (t_2 - t_1) \quad (9)$$

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

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

$$Ab.Cap = \frac{\sum_1^n M_{CO_{2_{ab}}}}{M_{MEA}} \quad (10)$$

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

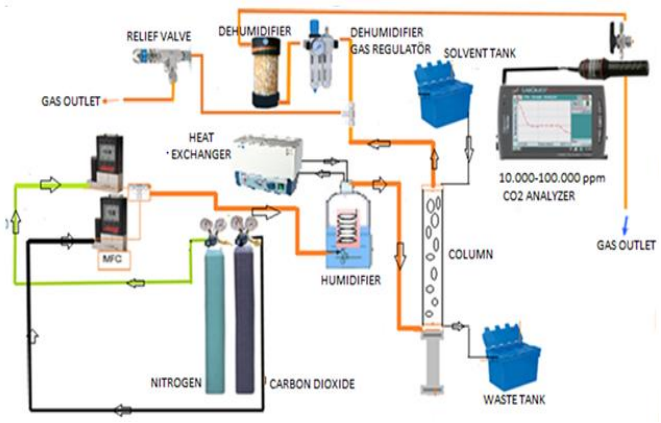

Figure 2. Experimental setup

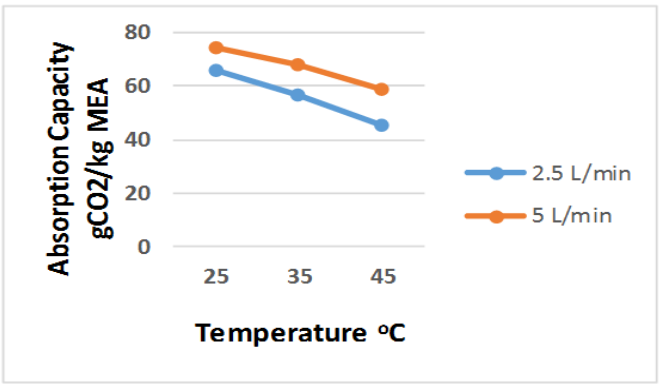

Figure 3. The effect of temperature on the absorption capacity (g CO_2/kg MEA)

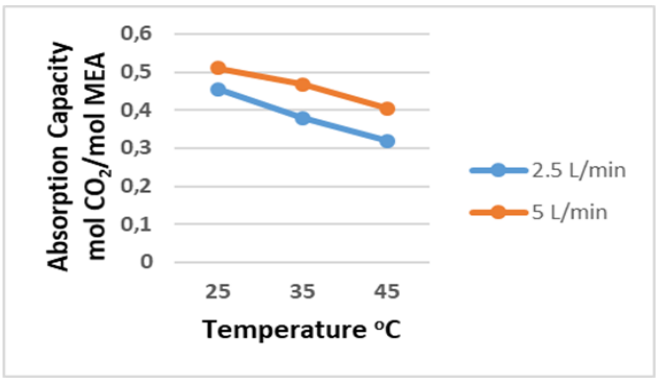

Figure 4. The effect of temperature on the absorption capacity (mol CO_2/mol MEA)

Chemicals

MEA was supplied by Sigma (Germany). N_2 (>99.99 %, 200 bar) and CO_2 (>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 CO_2 and N_2 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 CO_2 concentration was adjusted to 50,000 ppm using mass flow controllers. At the beginning of the experiment gas mixture was fed to the CO_2 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 CO_2 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

CO_2 absorption capacity is an essential feature for the successful CO_2 removal in a bubble column. The absorption capacity can be expressed in two units as mol of CO_2 absorbed per mol of amine solution and as g CO_2 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 CO_2/kg solvent and 0.456 mol CO_2/mol MEA decreases to 45.67 g CO_2/kg solvent and 0.32 mol CO_2/mol MEA with increasing temperature from 25 to 45 °C when using %20 MEA solution. This was due to the thermodynamics of the exothermic CO_2 absorption system that could cause reversible reactions when the temperature was too high. The increase in temperature could also increase the CO_2 vapor pressure over the solution that leads to decrease in the physical solubility of CO_2 in the solvent (Tan et al., 2012).

The absorption capacity of 74.71 g CO_2/kg MEA was obtained at 5 L/min gas flow rate, 25°C temperature and 20% solvent concentration. Similarly, Maneeintr et al. (2009) obtained 80g CO_2/kg MEA absorption capacity at 20% MEA and 40°C. Aronu et al. (2011) obtained 76 g CO_2/kg MEA absorption capacity at 3.0 M (%20) MEA, 40°C temperature and 15kpa CO_2 with using bubble column.

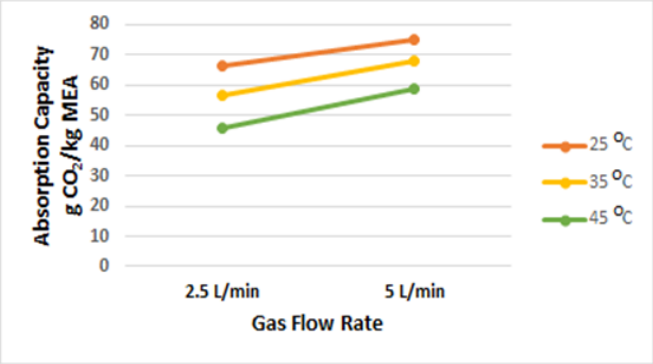

Figure 5. The effect of gas flow rate on the absorption capacity (g CO_2/kg MEA)

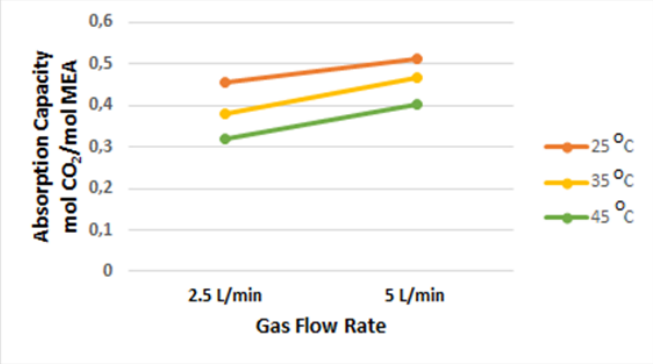

Figure 6. The effect of gas flow rate on the absorption capacity (mol CO_2/mol MEA)

The Effect of Gas Flow Rate on Absorption Capacity

The gas flow rate determines the detention time of the gas component and effects 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 CO_2 captured per solvent amount increases with increasing gas flow rate. The absorption capacity of 66.27 g CO_2/kg solvent and 0.456 mol CO_2/mol MEA was increased to 74.71 g CO_2/kg solvent and 0.51 mol CO_2/mol MEA when the gas flow rate increased from 2.5 L/min to 5 L/min. Conway et al. (2015) obtained 0.13 mol CO_2/mol MEA absorption capacity at 4M (%25) MEA and 3L/min gas flow rate with using wetted-wall column contactor.Aronu et al. (2011) obtained 0.516 mol CO_2/mol MEA absorption capacity at 2.5 M (%15) MEA, 40°C temperature and 4.84 kpa partial pressure of CO_2 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 CO_2 and water is strengthened and more CO_2 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 CO_2/kg solvent and mol CO_2/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 CO_2/kg solvent and 0.32 mol CO_2/mol MEA increased to 66.27 g CO_2/kg solvent and 0.456 mol CO_2/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 CO_2/kg solvent and 0.456 mol CO_2/mol MEA to 74.71 g CO_2/kg solvent and 0.51 mol CO_2/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 CO_2 / kg MEA and 0.51 mol CO_2/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 CO_2 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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Declaration of interest: The authors declare that they have no competing interests.

Ethics approval and consent to participate: Not applicable.

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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