Effect of CO₂ Concentration and Liquid to Gas Ratio on CO₂ Absorption from Simulated Biogas Using Monoethanolamine Solution

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Abstract. In industrial scale, removal of CO₂ by chemical absorption from raw biogas represents an imperative treatment and the cutting-edge technology towards improvement of its quality and heat value. In this work, CO₂ absorption studies were conducted in an absorption column packed with Sulzer metal gauze packing with simulated biogas absorbed using 30 wt. % of monoethanolamine (MEA) solution. Experimental works were conducted to determine the influence of different CO₂ concentrations in feed gas (30 % and 40 %) and L/G ratio (0.6 and 0.7) and subsequently assessed in terms of CO₂ absorption efficiency along the column. The results showed that 30 % CO₂ in feed gas has higher removal efficiency as compared to 40 % CO₂ with the ability to remove 94 % CO₂ during the process. In addition, the CO₂ absorption studied on the L/G ratio proved that CO₂ removal was improved at higher L/G ratio of 0.7.

1. Introduction

Biogas represents a renewable energy source that has good calorific value, produced under anaerobic condition from decomposition of organic matter. The principal components of biogas consist of methane (CH₄) (40 % – 75 %) and carbon dioxide (CO₂) (15 % – 60 %) with other trace gasses such as hydrogen sulphide (H₂S) [1]. CO₂ contained in the biogas is in terms of combustion, an inert gas. Thus, removal of CO₂ through biogas treatment is an essential process to improve its heat value. For enhancement of biogas quality, several CO₂ capture technologies such as adsorption [2], absorption [3] and membrane separation [4] may be implemented. Among these processes, chemical absorption stands out as the most well-established approach for CO₂ removal in industries due to its high removal efficiency, fast absorption rate and high product purity [5], [6]. Alkanolamines group of solvent has shown an effective absorption performance in CO₂ removal application. A primary amine, monoethanolamine (MEA) is a commercial solvent that has been widely used for the CO₂ absorption process which offers high reaction rate while being cost-effective [7].

The absorption performance in packed column is strongly influenced by the operating conditions. It has been reported for simulated flue gas treatment, removal efficiency is primarily affected by the CO₂ concentration in feed gas, followed by liquid flow rate (L) and gas flow rate (G). Under low pressure
conditions, a noticeable decrease in CO₂ removal efficiency was observed with increasing CO₂ concentrations [3 – 5]. A similar conclusion was reached by Hairul et al. [11] using natural gas (NG) containing 30 – 50 % CO₂ at high pressure conditions up to 4.04 MPa. In addition, for optimization of operating conditions and costs, the L/G ratios in the column have also been deemed essential for consideration [7, 8]. However, the influence of L/G ratios in the previous researches were mostly focused on the flue gas treatment of feed gas containing low CO₂ concentrations (1 – 15 %).

Hence, this study sought to understand the performance of CO₂ absorption from simulated biogas into MEA solution, conducted at relatively higher CO₂ concentrations (30 % and 40 %). In addition, the effect of L/G ratio towards the efficiency of CO₂ absorption was also studied via control of both flow rates entering the absorption column. The absorption performance was then quantified by degree of CO₂ removal (%) along the height of column.

2. Materials and methods

2.1. Chemical and gasses
Monoethanolamine (MEA) (99 % purity) was purchased from Merck, Germany. The CO₂ along with the natural gas (NG) which consists of CH₄ (97 %), CO₂ (2 %) and heavier hydrocarbon (1 %) were purchased from Air Product Malaysia and Petronas Dagangan Bhd, respectively. All chemicals and gasses for experimental works were used without additional purification.

2.2. Equipment and procedures
A fabricated packed absorption column with total height of 2.04 m and internal diameter of 0.046 m was used for all experiments. Sulzer metal gauze packing (Sulzer Chemtech Pte Ltd., Winterthur, Switzerland) with approximate surface area of 500 m²/m³ was packed in the column with 6 sampling points placed vertically at 0.34 m intervals.

Experimental setup of the process is as depicted in Figure 1. The simulated biogas was prepared as a mixture of NG and CO₂. Composition of the feed gas was determined by setting the mass flow controller of NG and CO₂ gases independently.

![Figure 1. The experimental set up for the absorption process.](image-url)
The mixed gas was pressurized up to 30 bar and kept in a gas vessel. From the bottom of the column, the simulated biogas was then introduced to flow upwards, controlled by a gas flow controller at a desired flow rate. Liquid phase was instead introduced from top of the column to travel downwards, thus establishing counter-current contact with the gas. Steady state condition of the absorption process was deemed reached upon operation for 30 minutes. Sampling was then conducted using CO$_2$-CH$_4$ IR Gas Analyzer (Fuji Electric Instrument, Japan) at each level of the absorption column under steady state condition. The CO$_2$-rich solvent was discharged from the bottom part of the absorption column and collected in a CO$_2$-rich solvent tank.

30 wt. % MEA solution with the simulated biogas containing either 30 % or 40 % CO$_2$ in NG gas acted as the liquid phase and gas phase, respectively. The CO$_2$ concentrations of 30 % and 40 % in NG were chosen to represent the intermediate concentration of raw biogas in real application. For process screening purpose, L/G ratios used were set at 0.6 and 0.7 by altering the gas flow rate while keeping constant the liquid flow rate. The experiments were conducted at operating pressure of 1.5 - 2 bar in the column. The absorption performance was finally quantified in terms of CO$_2$ removal percentage along the column and calculated using Equation (CO$_2$ removal (%) = $\frac{y_i - y_o}{y_i} \times 100\%$ (1)), where $y_i$ and $y_o$ represent the CO$_2$ mole fraction at the inlet and outlet of the column, respectively.

$$CO_2 \text{ removal (\%)} = \frac{y_i - y_o}{y_i} \times 100\%$$

3. Results and discussion

3.1. The influence of CO$_2$ concentration in feed gas on the CO$_2$ removal efficiency

The CO$_2$ removal at each level of the column at a fixed gas and liquid flow rate for both CO$_2$ concentrations is as depicted in Figure 2. From the graph, the feed gas containing 30 % CO$_2$ and 40 % CO$_2$ can be seen to have achieved 94 % and 82 % of CO$_2$ removal, respectively. Lower removal efficiency at higher CO$_2$ concentrations under the same operating conditions may be due to the increased CO$_2$ mole fraction to react with limited free active amines in the liquid phase [14]. Moreover, similar increasing trend of CO$_2$ removal was demonstrated by both CO$_2$ concentrations. The reactive area with highest removal can be seen in the middle section of the column in between 0.68 m and 1.36 m. In this section, the concentration of CO$_2$ in the gas phase decreases as it moves upward along the column due to absorption of several CO$_2$ molecules into the liquid phase. In the meantime, CO$_2$ loading in the solvent gradually increases as the liquid moving downwards and limits the reaction with CO$_2$ molecules in the gas phase. Hence, most reactions between amine and CO$_2$ molecules occurred in the middle section of the column with significant increment in the removal efficiency [15].

![Figure 2. Effect of different CO$_2$ concentrations in feed gas on CO$_2$ removal using MEA solution. (Gas flow rate = 24.63 kmol/m$^2$.hr; liquid flow rate = 3.75 m$^3$/m$^2$.hr; [MEA] = 30 wt.%; P = 1.5 bar)](image-url)
3.2 The influence of L/G ratio on the CO₂ removal efficiency

Figure 3 shows the effect of L/G ratio at 0.6 and 0.7 on the CO₂ absorption performance using 30 wt.% MEA solution. The CO₂ removal at 0.7 ratio was about 73%, which was 15% higher than 0.6 ratio. This observation was possibly due to higher availability of amine molecules at the higher ratio to react with CO₂ molecules, consequently leading to higher absorption efficiency. Subsequently, at lower L/G ratios (higher gas flow rate), shorter residence time of gas in the absorption column further limits the reaction between CO₂ molecules and amine solvent. The same observation was also demonstrated by Zeng et al. [8] and Kasikamphaiboon et al. [16].

![Figure 3. Effect of L/G ratio on CO₂ removal using MEA solution.](image)

Figure 3. Effect of L/G ratio on CO₂ removal using MEA solution. (Liquid flow rate = 3.75 m³/m².hr; [MEA] = 30 wt.%; [CO₂] = 40 %; P = 2 bar)

4. Conclusion

The CO₂ absorption process was carried out in a packed column using 30 wt.% MEA solution with the influence of CO₂ concentration and L/G ratio on the removal performance investigated. This study proved that the efficiency of MEA as an absorbent was enhanced at low CO₂ concentration (30 % CO₂) in the feed gas and the parametric study on the effect of L/G ratio at 0.7 was higher than that at 0.6. Hence, this study concludes that both CO₂ concentration in feed gas and L/G ratio significantly affect the absorption performance and therefore play an important role in improving the removal process.

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