Journal of Applied Science and Agriculture, 10(5) Special 2015, Pages: 205-209

AENSI Journals

JOURNAL OF APPLIED SCIENCE AND AGRICULTURE

ISSN 1816-9112

Journal home page: www.aensiweb.com/JASA

Effect of Operating Pressure on CO_2 Absorption from Natural Gas in Packed Absorption Column

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ARTICLE INFO

Article history:
Received 28 January 2015
Accepted 25 February 2015
Available online 6 March 2015

Keywords:
CO2 capture, Chemical absorption, Monoethanolamine (MEA), High pressure operation

ABSTRACT

Background: Carbon dioxide (CO2) removal from natural gas is one of the main steps in natural gas purification which usually been conducted at high pressure conditions. However, most CO2 absorption studies that use amine based solvents were conducted at atmospheric pressure. **Objective:** This study reports the effect of operating pressure on CO2 removal from natural gas using monoethanolamine (MEA) solvent in a packed absorption column. The effects operating pressure (0.1, 3.0 and 5.0 MPa) and comparison of liquid flow rate (2.89 and 3.61 m3/m2.h) which conducted at 0.1 and 5.0 MPa were evaluated in terms of CO2 removal (%). **Results:** The operating pressure had improved the performance of CO2 absorption in a packed absorption column to approximately 60% CO2 removal when the operating pressure was increased from 0.1 to 5.0 MPa. The effect of liquid flow rate studied at different operating pressures also proved that at higher operating pressures, the absorption of CO2 was superior compared to at lower pressure. **Conclusion:** The operating pressure had a significant impact on CO2 absorption in natural gas using MEA solvent in a packed absorption column.

To Cite This Article: Hairul Nazirah Abdul Halim and Azmi Mohd Shariff, Effect of Operating Pressure on CO2 Absorption from Natural Gas in Packed Absorption Column. ***J. Appl. Sci. & Agric.,*** *10(5): 205-209, 2015*

INTRODUCTION

Carbon dioxide (CO_2) contributes to approximately 60% of total greenhouse gas emissions and is the main contributor to global warming. The main sources of CO_2 emissions are from major industrial flue gases such as natural gas burning and coal-fired power plants, iron and steel, refinery, petrochemical and cement industries. Additionally, CO_2, which exists as an impurity in raw natural gas, must be removed to increases the heating value as well as to meet the pipeline gas quality and sale gas specifications. Furthermore, CO_2 will react with water in the form of moisture to produce carbonic acid which would accelerate corrosion in the pipelines and other process equipments. CO_2 must also be removed in order to prevent crystallization of CO_2 during cryogenic processes (liquefaction process).

Researchers are actively studying CO_2 removal at atmospheric conditions and exploring potential absorbents for CO_2 removal in the range of 1 to 15% from flue gases. However, the CO_2 concentration in some natural gas reservoirs can be very high. For example, it could be as high as 71% in the Natuna gas field, Indonesia (Suhartanto *et al*., 2001). In terms of process condition, the main difference between CO_2 absorption from flue gas and natural gas is the operating pressure. The process of purifying raw natural gas from natural gas reservoir is usually conducted at high pressure conditions while the same process can be conducted at atmospheric condition for flue gas treatment.

Various technologies are available for the removal of carbon dioxide from industrial and natural gases such as membrane technology, physical and chemical absorptions as well as adsorption and cryogenic separations. Among them, chemical absorption has been proven to be the most effective for industrial processes (Rao and Rubin, 2002). This process is based on chemical reactions between the absorbed substances (such as CO_2) and the absorbent. This process generally consists of i) an absorption section where CO_2 is removed from a gas stream by a liquid solvent and ii) a stripping section where a regenerator is used to strip out CO_2 from the solvent. The gas stream flows upwards into the absorption unit and is counter-currently in contact with the absorbent which would result in CO_2 being transferred from the gas stream to the liquid solvent. The CO_2 rich solvent exits at the bottom of the absorber column and is sent to the stripping section. After going through the regeneration process, the low CO_2 content solvent (lean solvent) will be recovered

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from the stripping section and re-enters the top of the absorber column.

In terms of flue gas treatment, researches in CO_2 removal at low concentrations (< 15%) by using alkanolamines have been conducted at atmospheric pressure (Aroonwilas and Tontiwachwuthikul, 1997, 1998), (Aroonwilas *et al*., 2001), (Setameteekul *et al*., 2008), (Dey and Aroonwilas, 2009), (Fu *et al*., 2012), Naami *et al*., 2012), (Sema *et al*., 2013), (Artanto *et al*., 2014). The most commonly used alkanolamines for this purpose are monoethanolamine (MEA), diethanolamine (DEA), methyldiethanolamine (MDEA) and 2-amino-2-methyl-1-propanol (AMP).

Mass transfer studies in packed absorption columns reported in terms of interfacial area (a) and volumetric liquid mass transfer coefficient (k_La) for high pressure operations of up to 1.2 MPa and 5.0 MPa was reported by Benadda *et al*. (1996). A selectivity study of H_2S and CO_2 removal from nitrogen gas (N_2) using MEA in a packed absorption column at 1.0 MPa was conducted by Godini and Mawla (2008). They discovered that by increasing the pressure and the ratio of molar flow rate of liquid/molar flow rate of gas (L/G), the absolute efficiency in H_2S and CO_2 absorptions was also increased. The removal of 75 vol% CO_2 in NG at 1.0 MPa using a mixed amines solvent, known as Stonvent-II in a pilot scale absorption column packed with IMTP No. 25 packing was reported by Tan *et al*. (2012). They discovered that the performance of liquid temperature at 35°C was better than at 45°C. However, the performance of these chemical solvents at different operating pressures, specifically for natural gas purification, is still limited and requires further researches.

In this paper, the performance of CO_2 absorption from a mixture of 30 – 40% CO_2 in NG using MEA solvent is reported in terms of CO_2 removal (%) along the height of the column. The percentage of CO_2 removal was determined by conducting experiments at various operating pressures at 0.1, 3.0 and 5.0 MPa. The comparison of liquid flow rates at 2.89 and 3.61 m^3/m^2.h were conducted at 0.1 and 5.0 MPa. In this study, experiments were conducted in a bench scale absorber packed with Sulzer Metal Gauze Packing.

Methodology:

Chemical:

Acros Brand supplied the monoethanolamine (MEA) (99% purity) used in this research while CO_2 (99.99% purity) was procured from Air Product Malaysia. Petronas Dagangan Bhd supplied the NG containing 97% CH_4, 2% CO_2 and 1% heavier hydrocarbon. All materials have been used without further purification.

Equipment and Procedures:

An absorption column of 2.040 m high and 0.046 m internal diameter (ID) was packed with Sulzer Metal Gauze Packing, sponsored by Sulzer Chemtech Pte Ltd, based in Winterthur, Switzerland. This structured packing has an approximately 500 m^2/m^3 surface area. Both the absorption column and the structured packing have been fabricated from the 316 stainless steel. Additionally, for ease of measuring gas concentrations during experiments, 6 sampling points were strategically placed at different levels along the column height.

Fig. 1 depicts the schematic diagram of the experimental set up. The first step prior to conducting experiments involved preparing the desired gas composition of CO_2 and NG. This step was conducted by setting the flow rate of CO_2 and NG to the desired values using the designated mass flow controllers. Next, the resultant gas mix was compressed and stored in a high pressure gas vessel. During experiments, this gas will enter from the bottom of the packed absorption column. Its flow rate will be controlled with a gas flow controller.

The operating pressures during experiments ranges between 0.1 to 5.0 MPa. The pressurized absorption column has a back pressure regulator at its outlet which controls the pressure. MEA, the liquid absorbent used in this study, was pumped into the column using a high pressure liquid pump. This pump controls its flow rate through the top of the column which will produce a counter-current contact with the gas mix. Thus, CO_2 in the gas mix will be absorbed by the liquid MEA. The treated gas will leave the column from the top while the CO_2 rich solution will be stored in a tank at the bottom of the column.

All experiments have been conducted until a steady-state condition has been achieved. CO_2 concentrations at the inlet, outlet and the 6 sampling points were measured using a CO_2–CH_4 IR gas analyzer from Fuji Electric Instrument. Samples of gas concentrations along the height of the column were collected starting from the lowest level to a higher level. Readings were recorded after each level has achieved a steady state.

Eq. 1 below was used to determine the percentage of CO_2 removal;

$$CO_2 \text{ removal} = \frac{y_b - y_a}{y_b} \times 100\% \qquad \text{(Eq. 1)}$$

y_a and y_b denote the mole fraction of CO_2 in the gas mixture at the column's outlet and inlet, respectively.

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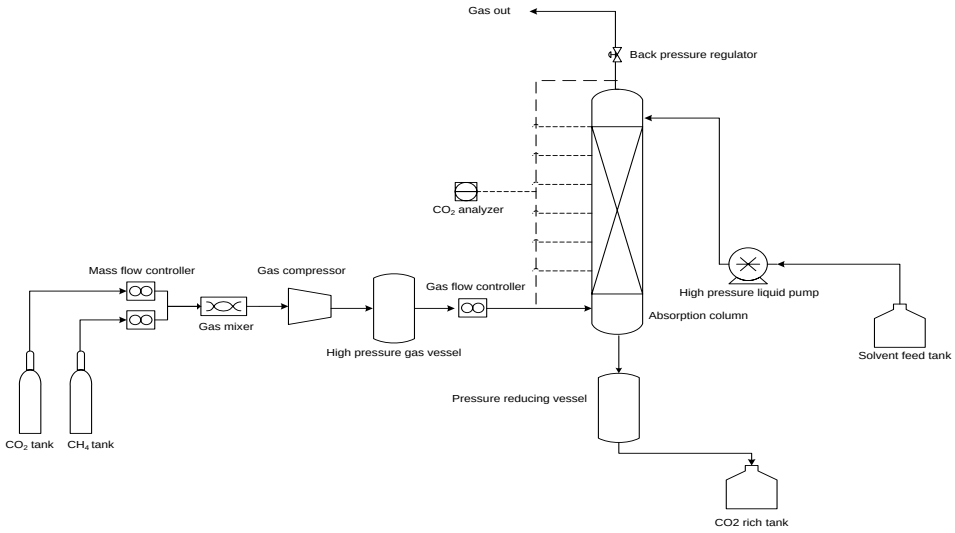

Fig. 1: Schematic diagram of the experimental set up.

RESULTS AND DISCUSSIONS

Effect of Operating Pressure on CO_2 Removal (%) in a Packed Absorption Column:

The effect of operating pressure, ranging from 0.1 to 5.0 MPa, was studied for a feed gas containing 30% CO_2 in NG. Fig. 2 shows the percentage of CO_2 removal using MEA along the height of the packed absorption column. High percentages of CO_2 removal have been observed for sampling points at the middle towards the top of the packed column. High mass transfer rate at the top of the column is expected due to the counter-current contact between the low CO_2 liquid absorbent and the CO_2-rich gas mix along the column. As the MEA flowed downwards, more CO_2 was absorbed, thus increasing CO_2 loading in the solvent. This condition decreases CO_2 removal at the lower section of the column due to the depleted amount of active amines available for the reaction.

Based on Fig. 2, CO_2 removal had increased to approximately 60% when the column pressure was increased from 0.1 to 5.0 MPa. At higher pressure, CO_2 partial pressure had increased while the equilibrium mole fraction in the gas (y*) had inversely varied with the increased total pressure. Thus, this condition leads to an increase in the mole-fraction driving force (y – y*) for separation. In the meantime, the decreasing liquid film resistance ($m/k_x a$) has resulted in a higher mass transfer performance at high pressure conditions (McCabe *et al*., 2005).

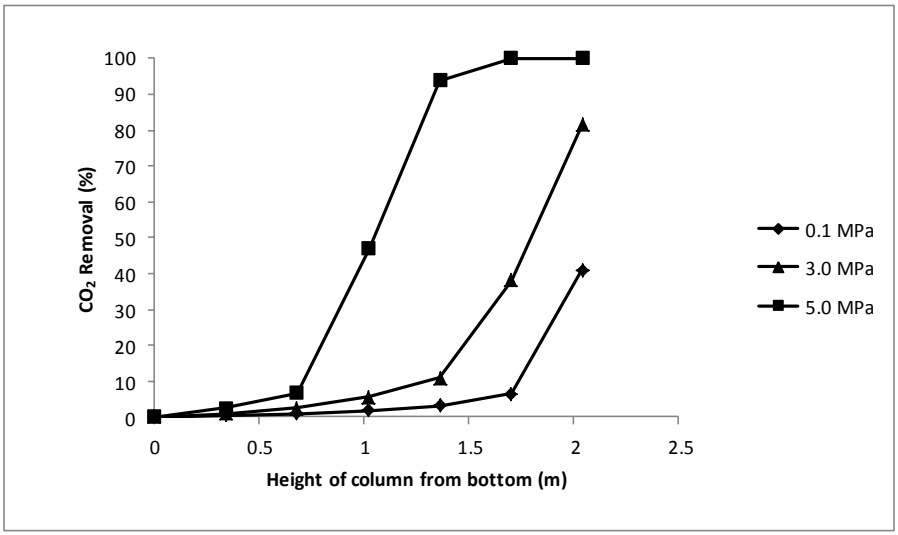

Fig. 2: Effect of column pressure on CO_2 removal (%) along the column height. (CO_2 concentration in NG = 30%, Gas flowrate = 30 kmol/m^2.h , MEA flow rate = 9.48 kmol/m^2.h, T = 27°C)

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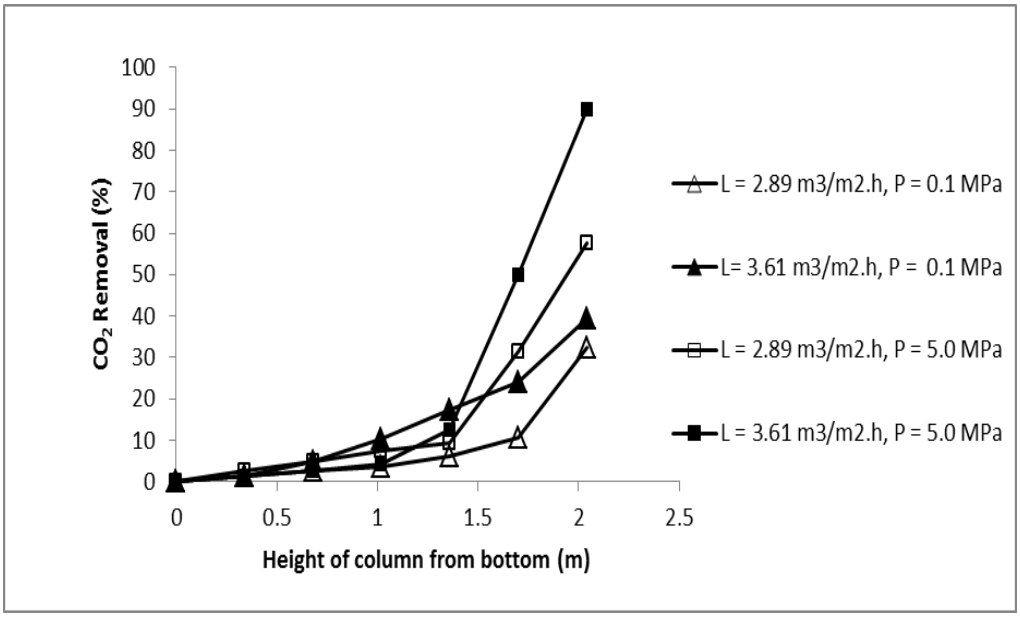

Fig. 3: Effect of liquid flow rate and operating pressure on CO_2 removal (%). (Gas flow rate: 33.18 kmol/m^2.h; 40% CO_2 in NG; [MEA] = 30 wt%; *T* = 27°C)

Effect of Liquid Flow Rate on CO_2 Removal (%):

The effect of liquid flow rate at 2.89 m^3/m^2.h and 3.61 m^3/m^2.h were conducted at the operating pressure of 0.1 MPa and 5.0 MPa. In these experiments, the gas flow rate and MEA concentration were fixed at 33.18 kmol/m^2.h and 30 wt%, respectively. The results are presented in Fig. 3.

Fig. 3 shows that CO_2 removal (%) had increased with the increase of liquid flow rate at the operating pressures of 0.1 MPa and 5.0 MPa. At 0.1 MPa, the percentage of CO_2 removal had increased by 7% when the liquid flow rate was increased from 2.89 m^3/m^2.h to 3.61 m^3/m^2.h. An increase of 32% was observed when the liquid flow rate was increased from 2.89 m^3/m^2.h to 3.61 m^3/m^2.h at 5.0 MPa operating pressure. This observation is the result of the enhanced reaction between the amine and CO_2 by the larger amount of free amine molecules at higher liquid flow rate. Furthermore, the decreased liquid film thickness had resulted in an increase in the liquid-side mass transfer coefficient. These observations corresponded well with CO_2 absorptions studied at atmospheric operations (Aroonwilas and Tontiwachwuthikul, 1997, 1998), (Aroonwilas *et al*., 2001), (Setameteekul *et al*., 2008), (Dey and Aroonwilas, 2009), (Maneeintr *et al*., 2010), (Zeng *et al*., 2011), (Fu *et al*., 2012), (Naami *et al*., 2012), (Sema *et al*., 2013).

Conclusions:

The effect of operating pressures (0.1, 3.0 and 5.0 MPa), on the removal of CO_2 from a gas mixture containing 30% CO_2 in NG, has been studied using a bench-scale absorption column packed with Sulzer Metal Gauze packing. The results have shown that the increased operating pressure had improved the performance of the CO_2 absorption process. The effect of liquid flow rate (2.89 and 3.61 m^3/m^2.h) conducted at 0.1 and 5.0 MPa indicated that CO_2 removal had increased with increasing liquid flow rate at both operating pressures.

ACKNOWLEDGMENTS

The authors would like to acknowledge the Research Centre for CO_2 Capture, University Technology PETRONAS (UTP) for funding this research; the scholarship support awarded to Hairul Nazirah Abdul Halim from the Ministry of Education Malaysia and Universiti Malaysia Perlis as well as Sulzer Chemtech Pte Ltd, Winterthur, Switzerland for sponsoring the Sulzer Metal Gauze Packing.

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