CARBON DIOXIDE ABSORPTION FROM BIOGAS USING PIPERAZINE-PROMOTED 2-AMINO-2-METHYL-1-PROPANOL BLENDED SOLUTION IN PACKED COLUMN

by

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LIST OF ABBREVIATIONS

1DMA2P	1-Dimethylamino-2-propanol
AEEA	Aminoethylethanolamine
AMP	2-amino-2-methyl-1-propanol
DEA	Diethanolamine
DEEA	N,N-diethylethanolamines
DETA	Diethylenetriamine
DIPA	Diisopropanolamine
DMEA	Dimethylethanolamine
EAE	Ethytaminoethanol
HMPD	4-Hydroxy-1-methylpiperidine
IR	Infrared
MDEA	Methyldiethanolamine
MEA	Monoethanolamine
MeOH	Methanol
NG	Natural gas
PZ	Piperazine
SHA	Sterically hindered amines
TEA	Triethanolamine

LIST OF SYMBOLS

C_{in}	Inlet CO_2 concentration, %
C_{out}	Outlet CO_2 concentration, %
E	Enhancement factor (dimensionless)
G	Gas flow rate, kmol/m^2·h
G_{CO_2}	Gas flow rate for CO_2 in gas, kmol/m^2·h
H	Henry's law coefficient, kPa/mol fraction
k_Ga_v	Individual volumetric mass transfer coefficient for the gas phase based on partial pressure driving force, kmol/m^3·h·kPa
K_Ga_v	Overall volumetric mass transfer coefficient for the gas phase based on partial pressure driving force, kmol/m^3·h·kPa
$\overline{K_Ga_v}$	Average overall volumetric mass transfer coefficient for the gas phase based on partial pressure driving force, kmol/m^3·h·kPa
$k_L^\circ a_v$	Mass transfer coefficient for the liquid phase without chemical reactions
k_xa_v	Individual volumetric mass transfer coefficient for the liquid phase based on unit mole fraction driving force, kmol/m^3·h·unit mole fraction
k_ya_v	Individual volumetric mass transfer coefficient for the gas phase based on unit mole fraction driving force, kmol/m^3·h·unit mole fraction
K_ya_v	Overall volumetric mass transfer coefficient for the gas phase based on unit mole fraction driving force, kmol/m^3·h·unit mole fraction
L_{amine}	Liquid flow rate for amine in liquid, m^3/m^2·h
L	Liquid flow rate, m^3/m^2·h
m	Slope of equilibrium curve
P	Total pressure, kPa
P_{CO_2}	Carbon dioxide partial pressure, kPa
$P^*_{CO_2}$	Carbon dioxide partial pressure at equilibrium, kPa
S	Cross-sectional area of column, m^2
T	Temperature, °C
V	Molar flow rate of gas, mol/h
vol.%	Volume percentage
wt.%	Weight percentage
y_a	Mole fraction of component A in the bulk gas at the column outlet, mol/mol
y_b	Mole fraction of component A in the bulk gas at the column inlet, mol/mol
Z_T	Packing height, m

Penyerapan Karbon Dioksida Daripada Biogas Menggunakan Larutan Campuran 2-Amino-2-Metil-1-Propanol Berpenggalak Piperazin Dalam Turus Terpadat

ABSTRAK

Biogas adalah sumber tenaga yang boleh diperbaharui yang umumnya terdiri daripada metana (CH_4) (40% hingga 75%) dan karbon dioksida (CO_2) (20% hingga 60%). Monoetanolamina (MEA) adalah penyerap mapan dalam penyerapan CO_2, namun prestasi penyingkirannya terbatas oleh limitasinya. Untuk mengatasi batasan penyerap amina tunggal, suatu penyerap amina campuran yang berpotensi; larutan campuran 2-amino-2-metil-1-propanol berpenggalak piperazin (PZ/AMP) menunjukkan prestasi cemerlang dalam menangkap CO_2 dalam keadaan tekanan separa CO_2 yang rendah (< 20 kPa) dan lebih sesuai untuk keadaan dalam rawatan gas serombong. Namun begitu, untuk aplikasi peningkatan mutu biogas, tekanan separa CO_2 yang lebih tinggi (> 20 kPa) harus dipertimbangkan kerana kepekatan CO_2 yang tinggi dalam gas suapan. Oleh kerana perilaku pemindahan jisim pada komposisi CO_2 yang lebih tinggi dalam gas suapan boleh mempengaruhi prestasi proses dalam turus terpadat, keberkesanan penyerap ini untuk menyingkirkan CO_2 pada tekanan separa CO_2 lebih tinggi daripada 20 kPa penting untuk diterokai. Oleh itu, dalam penyelidikan ini, penyerapan CO_2 daripada biogas simulasi dikaji menggunakan larutan campuran PZ/AMP yang berbeza dalam suatu sistem penyerapan pada tekanan 200 kPa. Prestasi penyerapan CO_2 dinilai berdasarkan nisbah larutan PZ terhadap AMP yang berbeza (0/30, 3/27, 5/25, 7/23, dan 9/21 wt.%/wt.%) dan ditanda aras dengan larutan MEA pada 30 wt.%. Kesan parameter proses terhadap penyerapan CO_2 ke dalam larutan PZ/AMP turut diteliti dan dibincangkan secara menyeluruh dari segi prestasi proses, termasuk tekanan separa CO_2 (20–110 kPa), kadar aliran gas (22.10–35.36 kmol/m^2·h), kadar aliran cecair (3.25–5.42 m^3/m^2·h), kepekatan bahan kimia (10–40 wt.%) dan suhu cecair salur masuk (30 ± 2 hingga 45 ± 2 °C). Data dibentangkan sebagai profil penyingkiran CO_2 di sepanjang turus dan dinilai dari segi kecekapan penyingkiran CO_2 (%) dan purata pekali pemindahan jisim isi padu keseluruhan dalam fasa gas ($\overline{K_G a_v}$). Didapati bahawa peningkatan kepekatan PZ dalam larutan AMP telah meningkatkan prestasi penyerapan CO_2 dan pemindahan jisim secara ketara, manakala larutan campuran 7 wt.% PZ/23 wt.% AMP menunjukkan prestasi setara dengan prestasi penyingkiran larutan MEA pada 30 wt.%. Peningkatan tekanan separa CO_2 dan kadar aliran gas telah mengurangkan prestasi penyingkiran CO_2 dari 100% ke 58% dan 72%. Nilai $\overline{K_G a_v}$ juga menurun dari 0.63 ke 0.039 kmol/m^3·h·kPa (tekanan separa CO_2) dan 0.317 to 0.092 kmol/m^3·h·kPa (kadar aliran gas). Sebaliknya, kadar aliran cecair dan kepekatan bahan kimia menunjukkan kesan positif terhadap prestasi penyingkiran CO_2. Hasil kajian ini menunjukkan bahawa peningkatan kadar aliran cecair dan kepekatan bahan kimia, meningkat kecekapan penyingkiran CO_2 dari 70% dan 33% ke 100%, serta peningkatan nilai $\overline{K_G a_v}$ dari 0.066 ke 0.389 kmol/m^3·h·kPa (kadar aliran cecair) and 0.024 ke 0.276 kmol/m^3·h·kPa (kepekatan bahan kimia). Manakala, suhu optimum cecair salur masuk diperhatikan pada 35 ± 2 °C dalam kajian ini, dengan penyingkiran 100% CO_2 dan $\overline{K_G a_v}$ bernilai 0.36 kmol/m^3·h·kPa. Keseluruhannya, larutan campuran PZ/AMP menunjukkan potensi besar untuk dikomersialkan dalam pengeluaran biogas perindustrian.

Carbon Dioxide Absorption from Biogas Using Piperazine-Promoted 2-Amino-2-Methyl-1-Propanol Blended Solution in Packed Column

ABSTRACT

Biogas is a renewable energy source that is mainly composed of methane (CH_4) (40%–75%) and carbon dioxide (CO_2) (20%–60%). Monoethanolamine (MEA) is an established absorbent in CO_2 absorption, yet its removal performances are challenged by its limitations. In order to overcome the limitations of individual amine absorbents, a potential blended amine solution; piperazine-promoted 2-amino-2-methyl-1-propanol (PZ/AMP) has a remarkable performance in capturing CO_2 at low CO_2 partial pressure conditions (< 20 kPa), and suited for the conditions in flue gas treatment. However, for biogas upgrading applications, higher CO_2 partial pressure (> 20 kPa) should be considered due to the presence of high CO_2 concentration in the feed gas. Since the mass transfer behaviour at higher CO_2 compositions in the feed gas can affect the process performance in a packed column, it is crucial to explore the effectiveness of the potential absorbent in removing CO_2 at higher than 20 kPa of CO_2 partial pressure. Therefore, in this research, CO_2 absorption from simulated biogas was investigated using different blends of PZ/AMP solution in an absorption system at 200 kPa. CO_2 absorption performance was evaluated at different ratios of PZ to AMP solution (0/30, 3/27, 5/25, 7/23, and 9/21 wt.%/wt.%) and was benchmarked with 30 wt.% of MEA solution. The effects of process parameters CO_2 absorption into PZ/AMP blended solution were also examined and thoroughly discussed, in terms of process performance, including CO_2 partial pressure (20–110 kPa), gas flow rate (22.10–35.36 kmol/m^2·h), liquid flow rate (3.25–5.42 m^3/m^2·h), chemical concentration (10–40 wt.%), and inlet liquid temperature (30 ± 2 to 45 ± 2 °C). The data were presented as CO_2 removal profiles along the column and evaluated in terms of CO_2 removal efficiency (%) and average overall volumetric mass transfer coefficient in the gas phase ($\overline{K_G a_v}$). Increased PZ concentration in AMP solution was found to have significantly increased CO_2 absorption and mass transfer performance, while the 7 wt.% of PZ/23 wt.% of AMP blend has a similar removal performance as 30 wt.% of MEA solution. Increased CO_2 partial pressure and gas flow rate in the process have been shown to decrease CO_2 removal performance from 100% to 58% and 72%, respectively. Meanwhile, the $\overline{K_G a_v}$ values were decreased from 0.63 to 0.039 kmol/m^3·h·kPa (CO_2 partial pressure) and 0.317 to 0.092 kmol/m^3·h·kPa (gas flow rate). In contrast, increased liquid flow rate and chemical concentration exhibited positive impacts on CO_2 removal performance. The results showed that increasing liquid flow rate and chemical concentration have increased the CO_2 removal efficiency from 70% and 33%, respectively, to a complete removal. Similarly, the $\overline{K_G a_v}$ values were increased from 0.066 to 0.389 kmol/m^3·h·kPa (liquid flow rate) and 0.024 to 0.276 kmol/m^3·h·kPa (chemical concentration). Meanwhile, the optimum inlet liquid temperature was observed at 35 ± 2 °C in this study, with a 100% CO_2 removal efficiency and 0.36 kmol/m^3·h·kPa of $\overline{K_G a_v}$ value. Overall, the PZ/AMP blended solution showed great potential to be commercialised in industrial biogas production.

CHAPTER 1 : INTRODUCTION

1.1 Background of Study

1.1.1 Biogas Constituents and Applications

Biogas is a renewable energy produced from the decomposition of organic materials by a series of microorganisms under anaerobic conditions. The constituents of raw biogas include methane (CH_4) (40%–75%) and carbon dioxide (CO_2) (20%–60%), with small concentrations of impurities, such as hydrogen sulphide (H_2S), hydrogen (H_2), nitrogen (N_2), ammonia (NH_3), and water vapour (Bharathiraja et al., 2018). This composition may also vary depending on the organic substrates used in the anaerobic digestion process for biogas production (Rafiee et al., 2021).

Different technologies have been implemented in the biogas sector to enhance the productivity and quality of raw gas. Upgraded biogas to biomethane is subsequently used in many applications with low environmental impact, such as substituting fossil fuel for transportation, gas fuel for electricity generation, and feedstock for various chemical industries. Additionally, biomethane combustion has the highest calorific value for energy generation among other transport biofuels, such as biodiesel, bioethanol, and biomethanol (Tabatabaei et al., 2020a).

1.1.2 CO_2 Removal in Biogas Upgrading Process

Biogas purification is the process of removing undesired gases, such as CO_2 and H_2S to increase its quality. These contaminants are highly undesirable in combustion systems because they can convert into highly corrosive and environmentally hazardous compounds. For instance, the combustion of biogas with high H_2S could release sulphur dioxide (SO_2) into the air that can cause acid rain (Tantikhajorngosol et al., 2019). CO_2 is also present in biogas as an inert gas, in terms of combustion. CO_2 can have a high impact on the calorific value of biogas, wherein the higher the composition of CO_2, the lower the calorific value of biogas. In this case, the presence of CO_2 can decrease the energy utilisation efficiency of the biogas. Therefore, biogas purification is a crucial process before any eventual utilisation.

To date, biogas upgrading technologies are available for commercial applications, including chemical and physical absorptions (Hu et al., 2018; Zhang et al., 2018), adsorption (Pioquinto et al., 2021; Creamer & Gao, 2016), membrane separation (Xu et al., 2018; Xu et al., 2019), and biological capture (Rachbauer et al., 2016). Among these technologies, chemical absorption using amine-based absorbents is the most applicable technology owing to its outstanding performance in producing high CH_4 purity and low CH_4 loses (Abdeen et al., 2016; Liang et al., 2015). Various absorbents have been used to intensify the absorption process. The most commonly used absorbents are aqueous solutions of alkanolamines that are classified into several amines groups, such as monoethanolamine (MEA), diethanolamine (DEA), diisopropanolamine (DIPA), and methyldiethanolamine (MDEA).

Monoethanolamine (MEA) is a primary amine that is known as a conventional absorbent for CO_2 capturing application due to its high reactivity, fast absorption rate, and low absorbent cost (Liao at al., 2017). However, the application of this conventional amine requires higher heat regeneration energy due to the formation of stable carbamate, leading to higher degradation rate. The corrosive nature of the absorbent, especially at high temperatures and concentrations, is also a disadvantage (Nwaoha et al., 2018). These limitations can directly affect absorption and regeneration efficiencies, which can consequently increase the operational cost.

An ideal absorbent is needed to achieve excellent performance in the CO_2 absorption process, with high absorption capacity, fast reaction rate, and low regeneration energy, corrosivity, and degradation rates (Muchan et al., 2017). However, the unique characteristics of single amines are lacking the criteria of an ideal absorbent for the CO_2 absorption process (Liang et al., 2016). Therefore, the main challenge is to find an alternative absorbent for the conventional absorbent. In the meantime, blended amine absorbents have gained interest for the removal process (Lee et al., 2020; Linget al., 2019; Singh et al., 2016). The synergistic effect of the blended amines on the absorption performance is achieved by combining the strengths of individual absorbents, while suppressing their limitations (Budzianowski, 2016).

Other than alkanolamines, sterically hindered amines (SHA), such as 2-amino-2-methyl-1-propanol (AMP), have been used as commercial absorbents for CO_2 absorption. The main interest in using AMP in CO_2 absorption is its equilibrium loading capacity, which is almost twice the capacity provided by MEA (Ooi et al., 2020; Koronaki et al., 2017). Theoretically, the CO_2 loading capacity of AMP is 1.0 mol CO_2/mol AMP due to the formation of bicarbonate that allows more CO_2–amine reaction to occur. Compared with MEA, the formation of unstable carbamate in the reaction is one of the highlights in using AMP as an absorbent, which would allow the regeneration process to achieve lower regeneration temperatures. Other attractive characteristics of AMP utilisation include lower degradation rate and corrosivity.

When compared to MDEA, a tertiary amine, both absorbents (AMP and MDEA) offered double CO_2 loading capacity (1.0 mol CO_2/mol amine), as opposed to MEA, a primary amine (0.5 mol CO_2/mol amine). In the reaction with CO_2, the formation of bicarbonate as the final product is also observed when using MDEA and AMP. However, in an experimental study on the performance of CO_2 removal using these absorbents, Jang et al. (2010) found that the best absorption efficiency of 81% was obtained using AMP, compared with 76% when using MDEA. Thus, the use of AMP as an absorbent in CO_2 removal process should be considered over MDEA for biogas upgrading applications.

As previously mentioned, AMP should be chosen as the primary absorbent in blended amine absorbents for its high CO_2 loading capacity, low regeneration energy, low degradation rate, and low corrosivity. However, AMP exhibits a slower reaction rate compared with MEA. For this reason, promoters should be added in AMP to accelerate the CO_2–amine reaction rate. Several research works have reported that piperazine-promoted (PZ) AMP is one of the promising blended amine absorbents for the application of CO_2 absorption from flue gas at low CO_2 partial pressure (Jahangiri & Hassankiadeh, 2019; Hassankiadeh & Jahangiri, 2018; Khan et al., 2016).

Piperazine is one of the prefered activator due to its advantageous solvent properties, fast reaction rate with CO_2 molecules, high equilibrium absorption loading capacity and less corrosion as compared to MEA. Besides, in Freeman et al. (2010) industrial study, they reported that regeneration energy of PZ is up to 20% lower than that of MEA. However, PZ solubility in amines is low which generally utilized in small amount for the absorption process. Consequently, the low concentration of PZ decreases its performance in terms of absorption capacity. Therefore, PZ is used as an promoter with other amines rather than as an individual solvent (Khan et al., 2016a).

1.2 Problem Statement

PZ/AMP blended solution has the potential to replace the conventional absorbents in commercial CO_2 absorption technology. The performance of PZ/AMP blended solution in removing low CO_2 concentration, specifically for flue gas treatment, was reported to be within 1–20 kPa of CO_2 partial pressure due to the presence of 1% to 15% of CO_2 concentration in the feed gas (Song et al., 2004). However, operations at higher than 20 kPa of CO_2 partial pressure are rarely discussed. Since 20% to 60% of CO_2 exists in biogas (Bharathiraja et al., 2018), biogas treatment at low pressure conditions should be performed at higher than 20 kPa of CO_2 partial pressure. Nonetheless, no research study has been conducted on the absorption performance of CO_2 using PZ/AMP blended solution at higher than 20 kPa of CO_2 partial pressure for biogas upgrading applications. Thus, the performance of PZ/AMP blended solution needs to be compared with the performance of MEA solution, as the benchmark absorbents in CO_2 absorption technology.

CO_2 removal performance is also believed to be influenced by the operating conditions in the packed column. The factors affecting the process performance in the packed column include CO_2 partial pressure, gas flow rate, liquid flow rate, chemical concentration, and inlet liquid temperature. Thus, it is crucial to evaluate the influence of these operating parameters on CO_2 absorption using PZ/AMP blended solution for maximum absorption performance.

While considering the mass transfer performance in the column, no significant change in the gas and liquid flow rates is expected during the absorption process at low CO_2 concentration in the feed gas (CO_2 dilute gas). However, in CO_2-rich gases, such as biogas, the mass transfer coefficient in the column will be affected by the dramatic changes in the gas and liquid flow rates along the column during the absorption process. Data of the average volumetric mass transfer coefficient ($\overline{K_G a_v}$) are important for upscaling and sizing of packed columns in biogas upgrading applications. Therefore, there is a need to study the influence of operating parameters on the efficiency of mass transfer in CO_2 absorption using PZ/AMP blended solution at higher than 20 kPa of CO_2 partial pressure for biogas upgrading applications.

1.3 Research Question

1. How efficient is the absorption process in capturing CO_2 from simulated biogas into PZ/AMP P blended solution compared with MEA and AMP solutions in a packed column?

2. How do the operating parameters affect CO_2 removal efficiency and mass transfer performance, in terms of CO_2 absorption from simulated biogas into PZ/AMP P blended solution in a packed column?

1.4 Objectives

1. To compare the performance of CO_2 removal from simulated biogas in a packed column using PZ/AMP blended solution and single amine solutions, namely, MEA and AMP.

2. To evaluate the influence of process parameters on CO_2 removal efficiency (%) and average overall volumetric mass transfer coefficient ($\overline{K_G a_v}$) using PZ/AMP blended solution at different CO_2 partial pressures, gas flow rates, liquid flow rates, chemical concentrations, and inlet liquid temperatures.

1.5 Scope of Study

In the current study, CO_2 absorption was conducted using an absorption column 0.046 m in internal diameter and operated at 200 kPa (2 bars). The internal section of the column was packed with 2.04 m of Sulzer metal gauze structured packing (Sulzer Chemtech Pte Ltd., Switzerland) at approximately 500 m^2/m^3 of packing surface area. The simulated biogas was prepared from a mixture of CO_2 and NG at the desired composition.

An industrial absorbent, MEA, was chosen as the benchmark absorbent in this experiment. The absorption of CO_2 into PZ/AMP blended solution was evaluated over different PZ to AMP mass fraction ratios (0/ 30, 3/27, 5/25, 7/23, and 9/21). Besides, the process parameters can have a high impact on the removal efficiency and operational costs of an industrial process. Thus, the removal performance in this study was observed in the range of 20 to 110 kPa of CO_2 partial pressure, mimicking the feed gas composition of the raw biogas. Most experiments were conducted at 80 kPa of CO_2 partial pressure to represent the intermediate composition of biogas at 40% of CO_2 concentration in raw biogas.

Moreover, poor handling of the operating parameters may possibly lead to several operational problems. Therefore, based on the dimension of the packed column used in this study, the gas flow rate was set within the 22.10–35.36 kmol/m^2·h range, while the liquid flow rate was set within the 3.25–5.42 m^3/m^2·h range in order to avoid channelling and flooding. Amine concentration and inlet liquid temperatures are also the important parameters to be studied in this process. Thus, the effect of different amine concentrations were analysed between 10 and 40 wt.%, while the effect of inlet liquid temperatures were investigated in the range of 30 ± 2 to 45 ± 2 °C. In addition, the inlet liquid temperature set up to 45 ± 2 °C due to the limitation of the system for heating up the feed solution and supplying the solution to the top of the column.

In this study, the CO_2 concentration profiles in gas phase along the column were measured from the bottom to the top of the column using an online CO_2–CH_4 infrared (IR) gas analyser. The experimental data were analysed and discussed in terms of CO_2 removal efficiency (%) and average volumetric mass transfer coefficient in the gas phase ($\overline{K_G a_v}$).

CHAPTER 2 : LITERATURE REVIEW

2.1 Biogas Upgrading Technologies

As a renewable energy source, biogas primarily consists of methane (CH_4) (40%–75%) with impurities, such as CO_2 (20%–60%) and trace gases, namely, hydrogen (H_2), ammonia (NH_3), hydrogen sulphide (H_2S), nitrogen (N_2), and water vapor (H_2O) (Bharathiraja et al., 2018). The compositions of biogas may vary depending on the organic substrates used in the anaerobic digestion process for biogas production (Tabatabaei et al., 2020b). Biogas upgrading is one of the essential processes to remove unwanted impurities resulting in methane-rich biogas with high calorific value (Miyawaki et al., 2021). The utilisation of methane-rich biogas is mainly for power generation and vehicle fuel in transportation. It is capable of replacing the use of fossil fuel as an energy resource. Recently, established biogas upgrading technologies for CO_2 and H_2S separation have become commercially available using adsorption, absorption, membrane separation, and cryogenic separation, as illustrated in Figure 2.1.

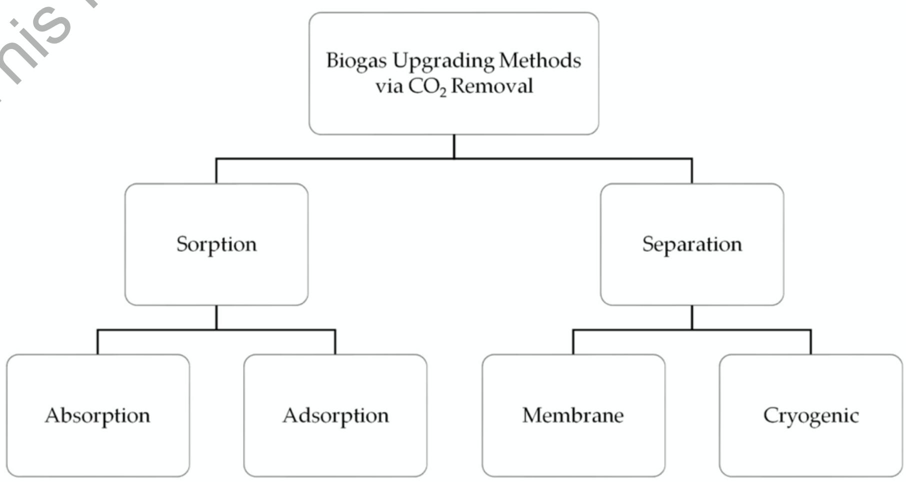

Figure 2.1: Biogas upgrading applications in CO_2 reduction (Adnan et al., 2019).