Songklanakarin J. Sci. Technol.
35 (6), 683-691, Nov. - Dec. 2013

SONGKLANAKARIN JOURNAL OF SCIENCE AND TECHNOLOGY
http://www.sjst.psu.ac.th

Original Article

Simultaneous removal of CO_2 and H_2S using MEA solution in a packed column absorber for biogas upgrading

Preecha Kasikamphaiboon[1], Juntima Chungsiriporn[1]*, Charun Bunyakan[1], and Wisitsree Wiyaratn[2]

[1] *Department of Chemical Engineering, Faculty of Engineering,*
Prince of Songkla University, Hat Yai, Songkhla, 90112 Thailand.

[2] *Department of Production Technology Education, Faculty of Industrial Education and Technology,*
King Mongkut's University of Technology Thonburi, Thung Khru, Bangkok, 10140 Thailand.

Received 27 December 2013; Accepted 6 August 2013

Abstract

Biogas production and utilization is an emerging alternative energy technology that has gained importance since the price of oil and gas has increased steadily over the last two decades. Biogas primarily consists of methane (CH_4) and carbon dioxide (CO_2) with smaller amounts of hydrogen sulfide (H_2S) and ammonia (NH_3). For many applications the quality of biogas has to be improved. The main parameters that may require removal in upgrading systems are CO_2 and H_2S. This work presents the study of simultaneous absorption of CO_2 and H_2S by Monoethanolamine (MEA) solution in a packed column. Simulated biogas containing 40% CO_2 and 60% N_2 and biogas generated from an anaerobic digestion plant were used as feed gas streams. The effects of gas flow rate, L/G ratio and absorbent concentration were investigated. The performance of the system was found to vary with process parameters. Increasing L/G ratio and MEA concentration causes the system efficiency to increase whereas increasing gas flow rate results in lower efficiency. An operating condition of L/G ratio of 83.3 ml/L, gas flow rate of 3 L/min and MEA concentration of 3 mol/L was found to remove more than 99.5% of CO_2 and H_2S from biogas. The volumetric overall mass-transfer coefficient (K_Ga_v) for CO_2 removal initially increases with increasing gas flow rate up to a certain value beyond which the coefficient becomes essentially constant. The K_Ga_v also increases with increasing L/G ratio throughout the range tested in this study.

Keywords: biogas, carbon dioxide, monoethanolamine, chemical absorption, packed column

---

1. Introduction

Biogas is a renewable fuel and an energy source that can be applied in many different settings. It is defined as a combustible gas mixture produced by the anaerobic digestion of biomass by bacteria and takes only a relatively short time to form. Biogas produced in anaerobic digestion plants is primarily composed of 55 to 65% of methane (CH_4) and 35-45% of carbon dioxide (CO_2) with smaller amounts of hydrogen sulfide (H_2S) and ammonia (NH_3) (Appels *et al.*, 2008). Methane (CH_4) combusts quite cleanly with hardly any soot particles or other pollutants, making it an almost clean fuel. Generally, biogas can be used directly to generate power, but the large volume of CO_2 reduces the heating value of the gas. For example, biogas containing 60% CH_4 has a calorific value of 21.5 MJ/Nm^3 while pure CH_4 has a calorific value of 35.8 MJ/Nm^3. The high CO_2 content in the biogas also increases compression and transportation costs and limits the economic feasibility for further use at the point of production. H_2S which is always present in biogas corrodes mechanical components in engine generator sets and vehicle engines. Moreover, sulfur dioxide (SO_2) produced from the combustion of H_2S can react with water in the atmosphere leading to acid rain problem. It is possible to upgrade or improve the quality

---

* Corresponding author.
Email address: juntima.c@psu.ac.th

of biogas by removing CO_2 and H_2S in order to enrich its CH_4 content up to the natural gas level, which is 75-98% (Vijay *et al.*, 2006). In Thailand, biogas resources are from industrial wastewater and live stock manure, which have a potential of 459 and 363 kilo tons of oil equivalent per year, respectively (DEDE, 2009). Therefore, upgrading the biogas allows for a wider variety of uses, either for heat and electricity production, or as a vehicle fuel.

Currently, technologies that could be used for CO_2 removal from gas include physical and chemical absorption, pressure swing adsorption (PSA), membrane separation, cryogenic separation and biological methane enrichment (Ryckebosch *et al.*, 2011). However, among these, CO_2 absorption using alkanolamines as chemical solvents appears to offer an interesting and practical alternative. In this technique, CO_2 is separated from a gas stream by passing the gas stream through a continuous scrubbing system consisting of an absorber and a desorber. After absorbing CO_2 in an absorber, the solvent is then sent to a desorber where the temperature is raised, resulting in CO_2 stripping from the solvent (solvent regeneration). The regenerated solvent is then returned to the absorber thereby creating a continuous recycling process. With the advantage of high efficiency and stability, packed column is widely used as the absorber and desorber in the CO_2 capture system (Yu *et al.*, 2010). In the existing industrial absorption processes, alkanolamines such as Monoethanolamine (MEA), Diethanolamine (DEA), Di-2-propanolamine (DIPA) and Methyldiethanolamine (MDEA) are commonly used (Mandal *et al.*, 2003). MEA is the most widely employed solvent for CO_2 absorption (Krumdieck *et al.*, 2008). Although new alkanolamines and alkanolamine mixes have been developed, MEA is still the preferred absorbent for CO_2 absorption. This is because MEA has a high reactivity, low solvent cost, low molecular weight (giving a high absorption capacity on a weight basis) and reasonable thermal stability (Ma'mun *et al.*, 2007). There are also several techniques to remove H_2S. Examples are absorption in water, chemical absorption in aqueous solutions and adsorption on solid adsorbents (Petersson and Wellinger, 2009). Over the years, a large amount of data on the CO_2 removal from flue gas and natural gas by using MEA has been published (e.g. deMontigny *et al.*, 2005, Godini and Mowla, 2008 and Lv *et al.*, 2012). However, there have been a few research papers applying such technology to biogas, which contains much higher CO_2 concentration than flue and natural gas. Moreover, a few works have focused on using MEA for simultaneous absorption of CO_2 and H_2S from biogas.

The objective of this work was, therefore, to evaluate the performance of a packed column for CO_2 removal from simulated biogas and for simultaneous removal of CO_2 and H_2S from biogas by using MEA solution as an absorbent. Effects of process parameters including gas flow rate, liquid to gas ratio (L/G) and concentration of MEA were investigated through a series of absorption experiments. The system performance was evaluated in terms of removal efficiency (%) and mass transfer coefficient.

2. Theory

2.1 Reaction schemes of CO_2 and H_2S with MEA

The chemistry of CO_2 reactions in amine solution is remarkably complex and cannot be said to be entirely understood. However, according to Choi *et al.* (2009), it has been generally accepted that the overall reaction of CO_2 with primary (RNH_2) such as MEA can be represented as in Reaction 1.

$$CO_2 + 2RNH_2 \leftrightarrow RNHCOO^- + RNH_3^+ \tag{1}$$

This mechanism comprises two steps, namely, formation of the CO_2-amine zwitterions as shown in Reaction 2, followed by base catalyzed deprotonation of this zwitterions as shown in Reaction 3:

$$CO_2 + RNH_2 \leftrightarrow RNH_2^+COO^- \tag{2}$$

$$RNH_2^+COO^- + B \leftrightarrow RNHCOO^- + BH^+ \tag{3}$$

where B is a base which could be amine, OH^- or H_2O.

The reaction of H_2S and MEA is reversible and instantaneous. H_2S reacts with MEA by proton transfer. The reaction (Al-Baghli *et al.*, 2001) is shown in Reaction 4.

$$RNH_2 + H_2S \leftrightarrow RNH_3^+ + HS^- \tag{4}$$

2.2 Mass transfer coefficient

The absorption performance of a packed column can commonly be presented as the volumetric overall mass-transfer coefficient (K_Ga_v). The determination of K_Ga_v is based on both the mass flux and material balance of the transferred CO_2 across the gas-liquid interface where the absorption takes place. The common equation used for calculating the K_Ga_v is Equation 5 (Aroowilas and Veawab, 2004):

$$K_Ga_v = \left(\frac{G_I}{P(y_{CO_2} - y^*_{CO_2})}\right)\left(\frac{dY_{CO_2}}{dZ}\right) \tag{5}$$

where G_I is the inert gas velocity in kmol/(m^2·h), P is the total pressure of the system in kPa, Z is the column height in m, y_{CO2} and $y^*_{CO_2}$ are the mole fraction of CO_2 in the gas stream and equilibrium mole fraction of CO_2, respectively, and y_{CO_2} is the mole ratio of CO_2 in the gas stream. The $y^*_{CO_2}$ term can be evaluated using solubility data. However, for CO_2 absorption into MEA solution, the $y^*_{CO_2}$ term can be assumed to be zero since the chemical reaction is fast (deMontigny *et al.*, 2005).

In this work, Equation 5 was rewritten according to Setameteekul *et al.* (2008) to represent the CO_2 concentrations in the gas entering and leaving the absorber ($Y_{CO_2,in}$ and $Y_{CO_2,out}$). The local mass-transfer driving force $P(y_{CO_2} - y^*_{CO_2})$ was replaced by a logarithmic mean, ΔP_{LM}. The mass-transfer equation was then written as in Equation 6:

$$K_G a_v = \frac{G_I (Y_{CO_2,in} - Y_{CO_2,out})}{\Delta P_{LM} Z} \tag{6}$$

where $$\Delta P_{LM} = P\left(\frac{(y_{CO_2} - y^*_{CO_2})_{in} - (y_{CO_2} - y^*_{CO_2})_{out}}{\ln((y_{CO_2} - y^*_{CO_2})_{in} / (y_{CO_2} - y^*_{CO_2})_{out})} \right)$$

3. Materials and Method

3.1 Materials

Aqueous solutions of monoethanolamine (MEA) used for CO_2 absorption were prepared by diluting the concentrated MEA (97% MEA), obtained from Fisher, with deionized water. N_2 and CO_2 gases with a purity of 99.99% and 99.97%, respectively, were used to produce simulated biogas at a concentration of 40% CO_2 and 60% N_2. Biogas used as feed gas stream in this work was obtained from the wastewater treatment plant of a local swine farm.

3.2 Experimental procedure

3.2.1 CO_2 absorption from simulated biogas

CO_2 absorption experiments were conducted in a laboratory-scale absorption unit of which a simplified flow diagram is given in Figure 1. The unit consisted of an acrylic absorption column (0.05 m in diameter and 1.00 m in height), CO_2 and N_2 gas cylinders, flow meters, mixing chamber and solvent tanks. The absorption column was packed with 6-mm raschig rings to a height of 0.70 m. The void fraction of the packing layer was 0.76 m^3/m^3 and the specific area was 625 m^2/m^3.

Each experimental run began by introducing N_2 and CO_2 gases from cylinders through flow meters at desired flow rates to produce simulated biogas at a concentration of 40% CO_2, which was fed to the bottom of the column. The concentration of CO_2 in the feed gas was checked and adjusted until the desired value was obtained. An aqueous MEA solution was pumped at a given flow rate to the top of the column and sprayed through the nozzle. After absorbing CO_2 and traveling through the column, the CO_2-rich solution was collected continuously in the liquid receiving tank. This operation was continued for at least 15 minutes to allow the system to reach steady state. At this point, CO_2 concentration of the treated gas stream was measured. The process parameters and the experimental conditions are listed in Table 1.

3.2.2 H_2S absorption from biogas

The H_2S absorption procedure was the same as described for the CO_2 absorption. Biogas from a local swine farm was used as feed gas instead of the simulated biogas. MEA solution at a concentration 3.0 mol/L was used as absorbent in the absorption unit. At steady state, the feed and treated gas streams were sampled and analyzed for CO_2 and H_2S concentrations in order to calculate system efficiencies.

All measurements were done in triplicate and the results were plotted and analyzed using ANOVA (analysis of variance) at 5% level.

3.3 Gas sample analysis and calculation

The concentrations of CO_2 and CH_4 in the feed and treated gas stream were analyzed by a gas chromatograph equipped with a TCD detector and Porapak Q packed column. The analysis of H_2S concentration in the gas stream was performed using the standard method given by AOAC (1990). The volumetric overall mass-transfer coefficient ($K_G a_v$) was calculated by Equation 6. For the simulated biogas treatment, the CO_2 removal efficiency (η_{CO_2}) of the system was determined by Equation 7 (Aroowilas and Veawab, 2004), and for the biogas treatment, the CO_2 removal efficiency (η_{CO_2}) and H_2S removal efficiency (η_{H_2S}) of the system were determined by Equation 8 and 9 (Lu *et al.*, 2006), respectively:

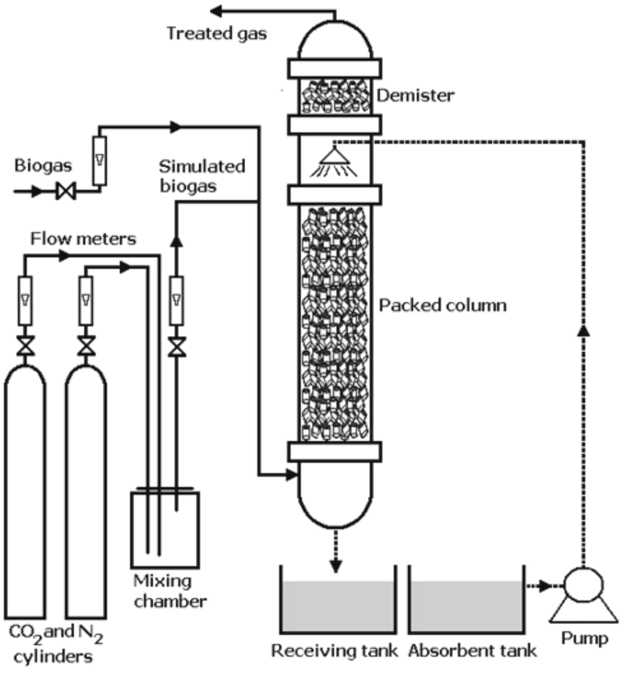


Figure 1. Schematic diagram of the gas absorption unit.

Table 1. Experimental operating conditions.

Parameter	conditions
Gas flow rate (L/min)	1.0-10.0
Liquid to gas ratio; L/G (ml/L)	16.7-83.3
MEA concentration (mol/L)	1.0-7.0
Liquid feed temperature (°C)	25
Absorption column pressure (kPa)	102.3

$$\eta_{CO_2} = \left[1 - \frac{y_{CO_2,out}}{y_{CO_2,in}}\left(\frac{1-y_{CO_2,in}}{1-y_{CO_2,out}}\right)\right] \times 100 \qquad (7)$$

$$\eta_{CO_2} = \left[1 - \frac{y_{CO_2,out}}{y_{CO_2,in}}\left(\frac{1-y_{CO_2,in}-y_{H_2S,in}}{1-y_{CO_2,out}-y_{H_2S,out}}\right)\right] \times 100 \qquad (8)$$

$$\eta_{H_2S} = \left[1 - \frac{y_{H_2S,out}}{y_{H_2S,in}}\left(\frac{1-y_{CO_2,in}-y_{H_2S,in}}{1-y_{CO_2,out}-y_{H_2S,out}}\right)\right] \times 100 \qquad (9)$$

where $y_{CO_2,in}$ and $y_{CO_2,out}$ denote mole fractions of gas phase CO_2 entering and leaving the absorption column, respectively, and $y_{H_2S,in}$ and $y_{H_2S,out}$ denote mole fractions of gas phase H_2S entering and leaving the absorption column, respectively.

4. Results and Discussion

4.1 CO_2 absorption from simulated biogas

4.1.1 Effect of gas flow rate

The effect of gas flow rate on CO_2 removal efficiency at three different MEA concentrations is shown in Figure 2. The CO_2 removal efficiencies of all MEA concentrations decrease as the gas flow rates increase. For example, when 3.0 mol/L MEA solution is used, the efficiency drops from about 100 to 81% as the gas flow rate increases from 1.0 to 10.0 L/min. It should be noted that the efficiency of the lowest MEA concentration (1.0 mol/L) is mainly affected by the increase in gas flow rate, decreasing dramatically from about 100 to 54% when the gas flow rate reaches 10.0 L/min. This is due to the limited amount of MEA in the solution to absorb CO_2. On the other hand, the CO_2 removal efficiency of the 5.0 mol/L MEA solution decreases gradually and is maintained at a level higher than 95% throughout the range of the gas flow rate.

4.1.2 Effect of liquid to gas ratio

Figure 3 shows the CO_2 removal efficiency as a function of L/G ratio at a gas flow rate of 3.0 L/min. The L/G ratio affects the CO_2 removal efficiency in the same pattern for all MEA concentrations used in this study. An increase in the L/G ratio results in an increase in efficiency. In the case of using 5.0 mol/L MEA solution, 96.2% of CO_2 in the gas stream is removed when an L/G ratio of only 16.7 ml/L is applied. Increasing the L/G ratio to 33.3 ml/L is enough causing the efficiency to reach a maximum of around 100%. Therefore, in practice, using L/G ratio greater than 33.3 ml/L may not be economical for this MEA concentration. For 3.0 mol/L MEA solution, removal efficiencies higher than 95% are obtained when the L/G ratios are in the range of 33.3-83.3 ml/L. However, for 1 mol/L MEA solution, an increase of L/G ratio from 16.7 to 33.3 ml/L cannot result in an efficiency of 95%, indicating insufficient amount of MEA to absorb CO_2 in the gas stream.

4.1.3 Effect of MEA concentration

The effect of MEA concentration was tested in this study to compare the performance of various absorbent concentrations at a gas flow rate of 6.0 L/min. The curve of CO_2 removal efficiencies of aqueous MEA solutions is shown in Figure 4. As can be seen that with an increase of absorbent concentrations from 1.0 to 7.0 mol/L, the CO_2 removal efficiency increases from 72.8 to 99.7%. This is due to the fact that increasing MEA concentration raises the molar ratio of MEA/CO_2, resulting in better CO_2 absorption and thus greater efficiency. This trend agrees well with the results obtained by Choi *et al.* (2009). However, it should be noted that the increasing rate of CO_2 removal efficiency decreases when MEA concentration is greater than 5.0 mol/L. This indicates

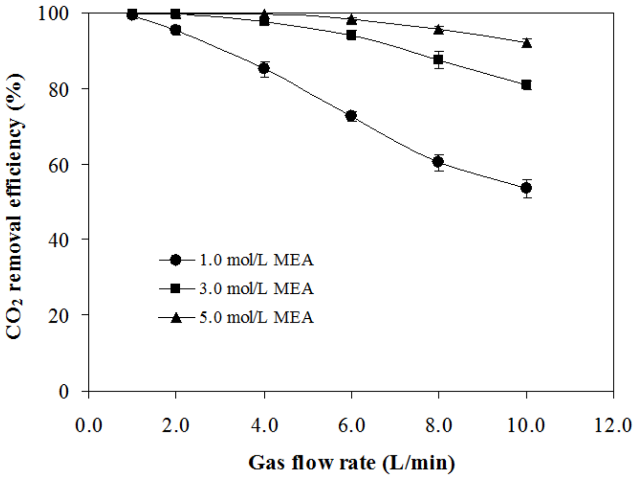


Figure 2. Effect of simulated biogas flow rate on CO_2 removal efficiency at a liquid flow rate of 0.15 L/min.

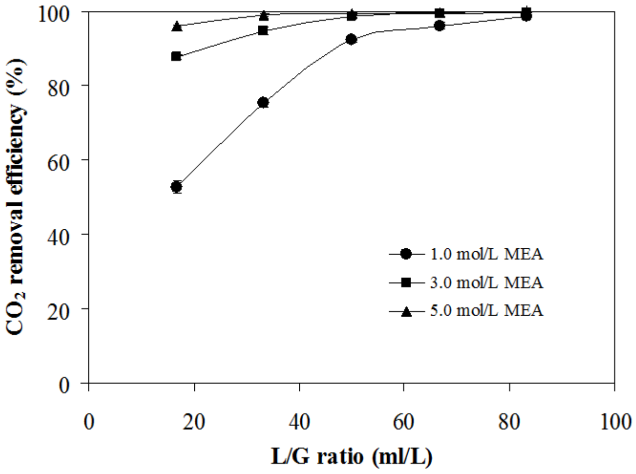


Figure 3. Effect of L/G ratio on CO_2 removal efficiency at a simulated biogas flow rate of 3.0 L/min.

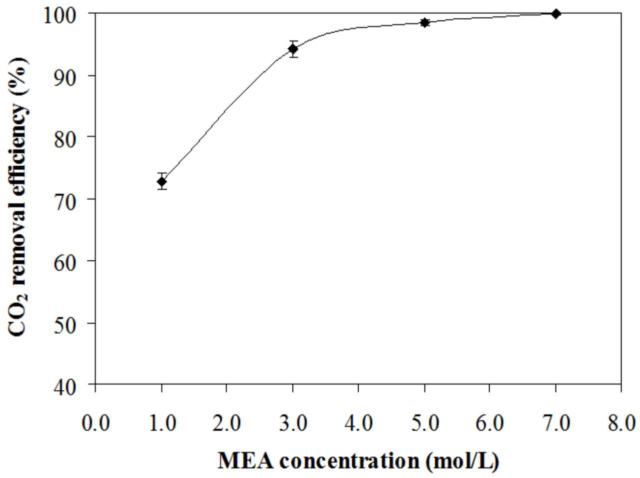


Figure 4. Effect of MEA concentration on CO_2 removal efficiency at a liquid flow rate of 0.15 L/min and a simulated biogas flow rate of 6.0 L/min.

that increasing concentration from 5.0 to 7.0 mol/L causes a slight increase in efficiency.

It is important to note that an increase in L/G ratio or solution flow rate leads to higher circulation and regeneration costs, while an increase in solution concentration is expensive and accelerate corrosion rates in steel vessels. Thus, maximizing these two operating parameters may not necessarily improve the overall system performance.

4.2 CO_2 and H_2S absorption from biogas

4.2.1 CO_2 absorption

From the analysis, the untreated biogas from the chosen swine farm consists of 61.5% of CH_4, 38.4% of CO_2 and 1,760 ppm of H_2S. These values are in the range of the typical biogas compositions found in literature (Appels *et al.*, 2008).The biogas was used as feed gas stream in this work. Plots of CO_2 removal efficiency and CH_4 outlet concentration against gas flow rate at 3.0 mol/L MEA solution are shown in Figure 5. It can be observed that the CO_2 removal efficiency decreases as the gas flow rate increases. Removal of CO_2 from the biogas results in a higher outlet CH_4 concentration. When gas flow rates in the range of 1.0-4.0 L/min are applied, the CO_2 removal efficiencies of 97.3-99.7% and the outlet CH_4 concentrations of 98.3-99.9% are obtained, corresponding to the outlet CO_2 concentrations of 0.1-1.7%. The system efficiency then drops as the gas flow rate is higher than 4.0 L/min. This indicates that increasing gas flow rate reduces contact time and increases the amount of CO_2 in the gas phase, thus resulting in lower efficiencies. Although the CH_4 content in the treated gas drops as the gas flow rate increases, using gas flow rates not higher than 6.0 L/min still maintains the outlet CH_4 concentrations at higher than 96.0% (less than 4.0% CO_2). In the application that uses biogas as vehicle fuel or NGV (natural gas for vehicle), the CO_2 and H_2S concentrations of the gas must be lower than 4% and 15 ppm, respectively (Deublein and Steinhauser, 2008).

The effect of L/G ratio on CO_2 removal efficiency was investigated by fixing the gas flow rate at 3.0 L/min and varying the L/G ratio in the range of 16.7-83.3 ml/L. Figure 6 shows that, at the lowest L/G ratio of 16.7 ml/L, the outlet CH_4 concentration is 90.8%. An increase in L/G ratio from 16.7 to 50.0 ml/L increases the efficiency to 99.1%. As the L/G ratio increases, the outlet CH_4 concentration increases. When L/G ratios of 50.0-83.3 ml/L are used, the efficiencies are almost as high as 100%.

4.2.2 H_2S absorption

Figure 7 shows the effect of gas flow rate on outlet H_2S concentration and removal efficiency. When a gas flow rate of 1.0 L/min is used, the H_2S concentration decreases from 1,760 ppm at the inlet to lower than 10 ppm at the outlet,

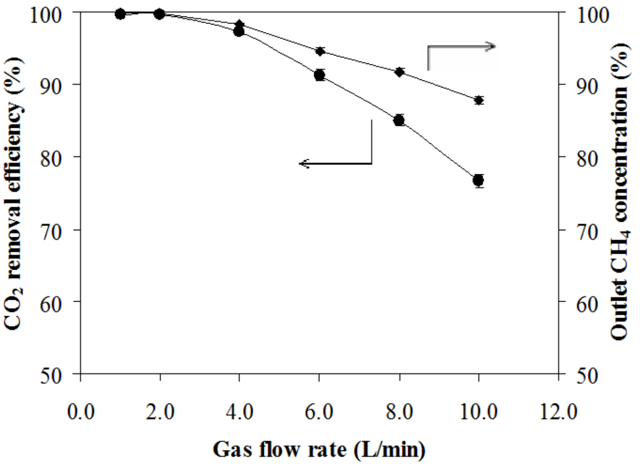


Figure 5. Effect of biogas flow rate on CO_2 removal efficiency at a liquid flow rate of 0.15 L/min.

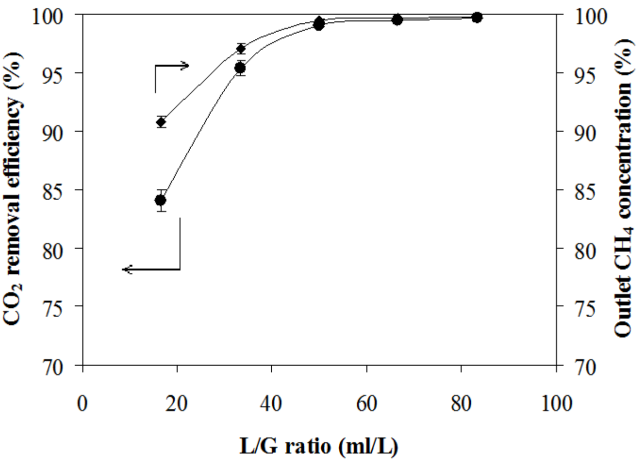


Figure 6. Effect of L/G ratio on CO_2 removal efficiency at a biogas flow rate of 3.0 L/min.

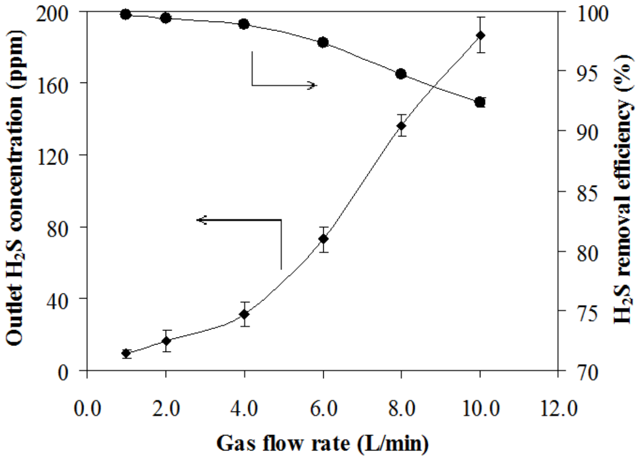


Figure 7. Effect of biogas flow rate on outlet H_2S concentration and removal efficiency at a liquid flow rate of 0.15 L/min.

corresponding to 99.7% of removal efficiency. Increasing gas flow rate from 1.0 to 10.0 L/min causes the outlet H_2S concentration to rise gradually, resulting in a decrease in efficiency. Despite the fact that the H_2S removal efficiency decreases as the gas flow rate increases, the efficiency is still higher than 95% as long as the gas flow rate is not higher than 8.0 L/min. However, as it was mentioned earlier, the requirement for using biogas as vehicle fuel is that the H_2S concentration of the gas must be lower than 15 ppm. Therefore, from Figure 6, the suitable gas flow rate should not be higher than 2.0 L/min.

Figure 8 shows outlet H_2S concentrations and H_2S removal efficiencies at different L/G ratios. At an L/G ratio of only 16.7 ml/L, the H_2S concentration is reduced from 1,760 to 76 ppm, corresponding to 97.1% of removal efficiency. Increasing L/G ratio decreases H_2S concentration. When an L/G ratio of 83.3 ml/L is used, a H_2S concentration as low as 10 ppm (99.7% efficiency) is obtained.

4.3 Mass transfer coefficient for CO_2 absorption

Beside the percent removal efficiency, the absorption performance of a packed column can be presented as the volumetric overall mass-transfer coefficient (K_Ga_v). The K_Ga_v coefficient is a combination of three contributions associated with mass transfer, i.e. thermodynamics, kinetics of the CO_2 absorption system and the hydrodynamic nature of the absorption equipment. Figure 9a shows the variation of the K_Ga_v as a function of simulated biogas flow rate for three different MEA concentrations. It is apparent that the gas flow rate affects the K_Ga_e but only to a certain point. The K_Ga_v increases initially with increasing gas flow rate up to around 6.0 L/min (183 $m^3/m^2 h$) beyond which the K_Ga_v tends to remain constant. This behavior illustrates the gas-phase controlled mass transfer taking place within the low range of the gas flow rate (less than 6.0 L/min). As the gas flow rate increases the K_Ga_v increases simultaneously to a point where the liquid-phase mass transfer takes over and becomes the main controlling factor for CO_2 absorption. A similar trend has been reported by Javed *et al.* (2010) and Kuntz and Aroonwilas (2009).

The effect of biogas flow rate on the K_Ga_v is shown in Figure 9b. The K_Ga_v increases significantly from 0.10 to 0.23 $kmol/m^3.h.kPa$ when the gas flow rate increases from 1.0 to 4.0 L/min beyond which the K_Ga_v becomes almost constant. This trend is similar to that of the 3.0 mol/L MEA solution in Figure 9a. Moreover, the values of the K_Ga_v for the simulated gas (Figure 9a) and for the biogas (Figure 9b) are almost the same. This indicates that, with the relatively very low concentration (less than 0.2%) compared to the CO_2 concentration in the feed gas stream, the H_2S does not affect the CO_2 absorption performance of the system.

The effect of L/G ratio on the K_Ga_v for the simulated biogas treatment is shown in Figure 10a. It is obvious that the K_Ga_v increases with increasing L/G ratio and this holds true for the entire range of L/G ratio tested. This is because the increase in L/G ratio or liquid flow rate results in the spread of liquid on the packing surface and more effective interfacial area between liquid and gas in the packing is formed producing a higher overall mass transfer to take place. Figure 10a also shows that the increase in K_Ga_v is observed for the entire range of MEA concentration tested. Previous study on the absorption of CO_2 into potassium carbonate (K_2CO_3) solutions (Zhao *et al.*, 2011) in a packed column has also revealed a similar trend of increasing K_Ga_v with increasing liquid flow rate. Figure 10b shows the effect of L/G ratio on the K_Ga_v for the biogas treatment. It can be seen that as L/G ratio increases, the K_Ga_v increases. This trend is similar to that in Figure 10a and the K_Ga_v values in the two figures at the same condition are almost the same.

The K_Ga_v values obtained from this study and from various references using a packed column technology for CO_2 absorption are compared in Table 2. It can be seen that most studies were carried out at relatively low CO_2 concen-

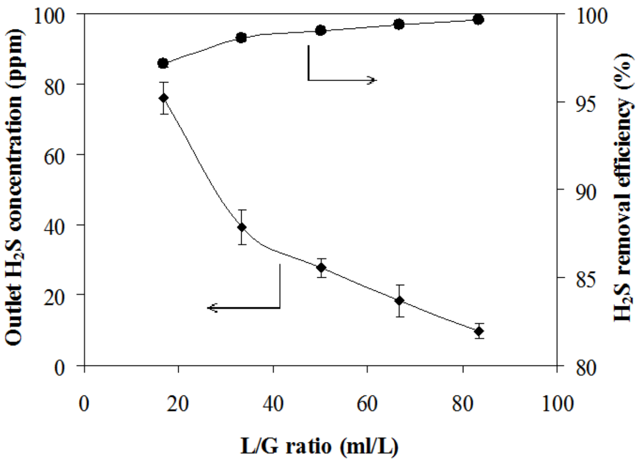
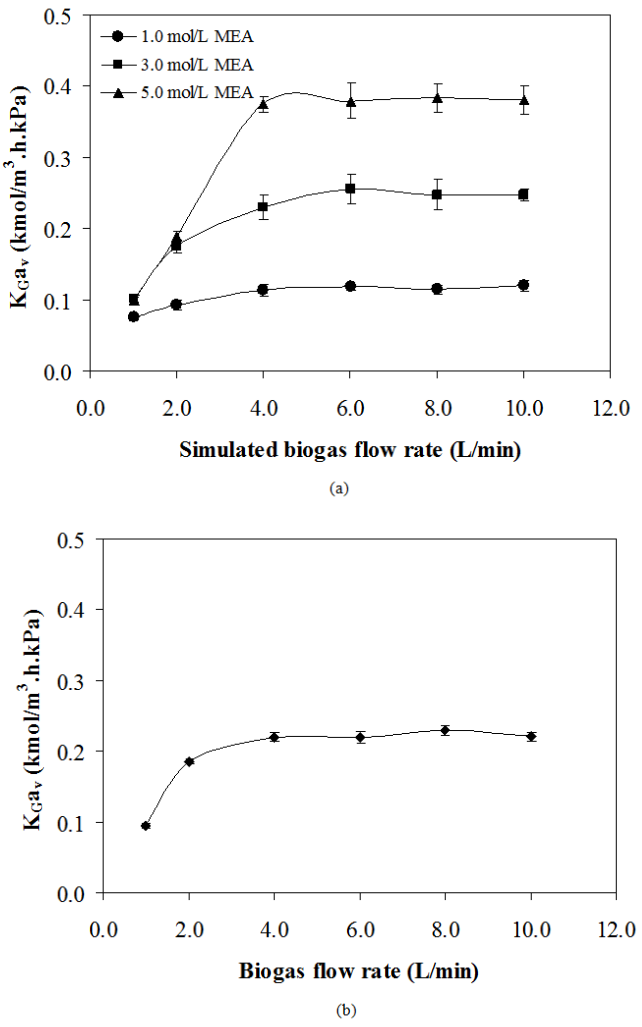


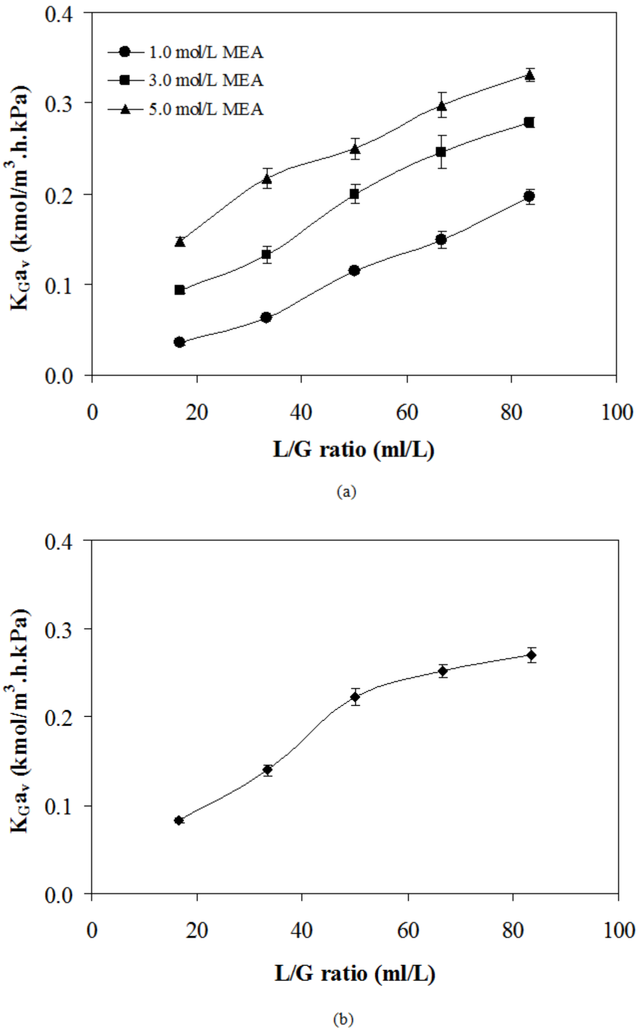
Figure 8. Effect of L/G ratio on outlet H_2S concentration and removal efficiency at a biogas flow rate of 3.0 L/min.



(a)

(b)

Figure 9. Effect of (a) simulated biogas flow rate and (b) biogas flow rate on K_Ga_v at a liquid flow rate of 0.15 L/min and an MEA concentration of 3.0 mol/L.



(a)

(b)

Figure 10. Effect of L/G ratio on K_Ga_v for (a) simulated biogas and (b) biogas treatment at a gas flow rate of 3.0 L/min and an MEA concentration of 3.0 mol/L.

Table 2. Comparison of K_Ga_v values reported in literature.

References	Gas composition	absorbents	K_Ga_v(kmol/m^3.h.kPa)
This work	40% CO_2, 60% N_2	1-5 mol/L MEA	0.1-0.4
	38% CO_2, 62% N_2	3 mol/L MEA	0.1-0.3
Kuntz and Aroonwilas (2009)	15% CO_2, 85% air	5 mol/L MEA	0.4-0.5
deMontigny *et al.* (2005)	8.4% CO_2, 91.6% air	2 mol/L MEA	1.1-1.3
Dey and Aroonwilas (2009)	5% CO_2, 95% N_2	5 mol/L MEA	1.3
	15% CO_2, 85% N_2		0.4
Aroonwilas and Tontiwachwuthikul (1998)	3-10% CO_2, 90-97% air	1.1 mol/L AMP	0.6-0.7
	15% CO_2, 85% air	3 mol/L AMP	1.0
Zeng *et al.* (2012)	5-15% CO_2, 85-95% N_2	1.2-4.5 mol/L Ammonia	0.1-0.4
Aroonwilas *et al.* (1999)	15% CO_2, 85% air	1.5 mol/L MEA	0.9
		1.5 mol/L NaOH	0.7

trations of about 15% or less, compared to that found in typical biogas (higher than 35%). Various chemicals used as absorbents include MEA, AMP (2-amino-2-methyl-1-propanol), Ammonia and NaOH (Sodium Hydroxide). The results from this study appear to be slightly different from those in the literature. With respect to MEA, the K_Ga_v values obtained from this study are observed to be lower than those from deMontigny *et al.* (2005) and Aroonwilas *et al.* (1999). This is probably due to the fact that they used lower CO_2 concentrations. Dey and Aroonwilas (2009) have found that K_Ga_v decreases with the increasing CO_2 concentration. This can be seen in Table 2, because when the CO_2 concentration is raised from 5 to 15%, the K_Ga_v drops from 1.3 to 0.4 kmol/m^3.h.kPa at the MEA concentration of 5.0 mol/L.

Compared to other techniques (Petersson and Wellinger, 2009; Scholz *et al.*, 2013), results obtained in this study show high efficiencies of simultaneous removal of CO_2 and H_2S from simulated biogas and biogas using MEA solution in a packed column. At suitable conditions, the system can effectively reduce CO_2 and H_2S concentration in the feed gas streams to lower than 1% and 10 ppm, respectively, with outlet CH_4 content higher than 99%. However, as mentioned earlier, gas absorption using alkanolamines as chemical solvents usually consists of two units, one for absorption and the other for solvent regeneration. Energy consumption for solvent regeneration is an important parameter that must be known for design and operation and economic analysis. Thus, further work dealing with CO_2 and H_2S absorption with the system equipped with a regenerating unit may be required.

5. Conclusions

Chemical absorption by MEA solution for simultaneously removing CO_2 and H_2S from biogas in a laboratory-scale packed column has been studied. Gas flow rate, liquid to gas ratio and MEA concentration are very important parameters affecting the process performance. Increasing L/G ratio and MEA concentration results in higher system efficiency whereas increasing gas flow rate cause the efficiency to decrease. For the simulated biogas treatment, the CO_2 removal efficiencies higher than 95% are obtained when using gas flow rates not higher than 6 L/min and L/G ratios of 33.3-50.0 ml/L. Results from the study also show that the CO_2 removal efficiency obtained from the biogas treatment is the same as that of the simulated biogas treatment at the same condition. The suitable condition that provides more than 99.5% of efficiency for CO_2 and H_2S removal from biogas is a gas flow rate of 3.0 L/min and an L/G ratio of 83.3 ml/L, at 3.0 mol/L of MEA solution. The volumetric overall mass-transfer coefficient (K_Ga_v) for CO_2 removal initially increases with increasing gas flow rate, but at higher flow rates it is essentially constant. The K_Ga_v also increases with L/G ratio and MEA concentration.

Acknowledgments

The authors are grateful to the Ministry of Science and Technology for providing funding for this research. The authors also thank the Department of Chemical Engineering, Faculty of Engineering and The Graduate School of Prince of Songkla University for their support.

References

Al-Baghli, N.A., Pruess, S.A., Yesavage, V.F. and Selim, M.S. 2001. A rate-based model for the design of gas absorbers for the removal of CO_2 and H_2S using aqueous solutions of MEA and DEA. Fluid Phase Equilibria. 185, 31–43.

Appels, L., Baeyens, J., Degrève, J. and Dewil, R. 2008. Principles and potential of the anaerobic digestion of waste-activated sludge. Progress in Energy and Combustion Science. 34, 755–781.

Aroowilas, A. and Tontiwachwuthikul, P. 1998. Mass transfer coefficients and correlation for CO_2 absorption into 2-Amino-2-methyl-1-propanol (AMP) using structured packing. Industrial & Engineering Chemistry Research. 37, 569-575.

Aroowilas, A., Veawab, A. and Tontiwachwuthikul, P. 1999. Behavior of the Mass-Transfer Coefficient of Structured Packings in CO_2 Absorbers with Chemical Reactions. Industrial & Engineering Chemistry Research. 38, 2044-2050.

Aroowilas, A. and Veawab, A. 2004. Characterization and Comparison of the CO_2 Absorption Performance into Single and Blended Alkanolamines in a Packed Column. Industrial & Engineering Chemistry Research. 43, 2228-2237.

Association of Official Analytical Chemists (AOAC). 1990. Official methods of analysis of the association of official analytical chemists 15th ed, AOAC, Arlington, USA.

Choi, W.-J., Seo, J.-B., Jang, S.-Y., Jung, J.-H. and Oh, K.-J. 2009. Removal characteristics of CO_2 using aqueous MEA/AMP solutions in the absorption and regeneration process. Journal of Environmental Sciences. 21, 907–913.

Department of Alternative Energy Development and Efficiency (DEDE). 2009. Thailand Alternative Energy Situation 2009, Department of Energy Development and Promotion (DEDP), Bangkok, Thailand.

Deublein, D. and Steinhauser, A. 2008. Biogas from Waste and Renewable Resources: An Introduction, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany.

Dey, A. and Aroonwilas, A. 2009. CO_2 absorption into MEA-AMP blend: mass transfer and absorber height index. Energy Procedia. 1, 211-215.

deMontigny, D., Tontiwachwuthikul, P. and Chakma, A. 2005. Comparing the Absorption Performance of Packed Columns and Membrane Contactors. Industrial & Engineering Chemistry Research. 44, 5726-5732.

Godini, H.R. and Mowla, D. 2008. Selectivity study of H_2S and CO_2 absorption from gaseous mixtures by MEA in packed beds. Chemical Engineering Research and Design. 86, 401-409.

Javed, K.H., Mahmud, T. and Purba, E. 2010. The CO_2 capture performance of a high-intensity vortex spray scrubber. Chemical Engineering Journal. 162, 448-456.

Krumdieck, S., Wallace, J. and Curnow, O. 2008. Compact, low energy CO_2 management using amine solution in a packed bubble column. Chemical Engineering Journal. 135, 3-9.

Kuntz, J. and Aroowilas, A. 2009. Mass-transfer efficiency of a spray column for CO_2 capture by MEA. Energy Procedia. 1, 205-209.

Lu, J.G., Zheng, Y.F., and He, D.L. 2006. Selective absorption of H_2S from gas mixtures into aqueous solutions of blended amines of methyldiethanolamine and 2-tertiarybutylamino-2-ethoxyethanol in a packed column. Separation and Purification Technology. 52, 209–217.

Lv, Y., Yu, X., Tu, S.T., Yan, J. and Dahlquist, E. 2012. Experimental studies on simultaneous removal of CO_2 and SO_2 in a polypropylene hollow fiber membrane contactor. Applied Energy. 97, 283–288.

Ma'mun, S., Svendsen, H.F., Hoff, K.A. and Juliussen, O. 2007. Selection of new absorbents for carbon dioxide capture. Energy Conversion and Management. 48, 251–258.

Mandal, B.P., Biswas, A.K., Bandyopadhyay, S.S. 2003. Absorption of carbon dioxide into aqueous blends of 2-amino-2-methyl-1-propanol and diethanolamine. Chemical Engineering Science. 58, 4137 – 4144.

Petersson, A. and Wellinger, A. 2009. Biogas upgrading technologies-developments and innovations, IEA Bioenergy, Malmö, Sweden.

Ryckebosch, E., Drouillon, M. and Vervaeren, H. 2011. Techniques for transformation of biogas to biomethane. Biomass and Bioenergy. 35, 1633-1645.

Scholz, M., Melin, T. and Wessling, M. 2013. Transforming biogas into biomethane using membrane technology. Renewable and Sustainable Energy Reviews. 17, 199–212.

Setameteekul, A., Aroonwilas, A. and Veawab, A. 2008. Statistical factorial design analysis for parametric interaction and empirical correlations of CO_2 absorption performance in MEA and blended MEA/MDEA processes. Separation and Purification Technology. 64, 16–25.

Vijay, V.K., Chandra, R., Parchuri M., Subbarao, V. and Kapdi, S.S. 2006. Biogas purification and bottling into CNG cylinders: producing Bio-CNG from biomass for rural automotive applications. Proceeding of the 2nd Joint International Conference on "Sustainable Energy and Environment (SEE 2006)". Bangkok, Thailand, November 21-23, 2006, 1-6.

Yu, Y.S., Li, Y., Lu, H.F., Yan, L.W., Zhang, Z.X., Wang, G.X. and Rudolph V. 2010. Multi-field synergy study of CO_2 capture process by chemical absorption. Chemical Engineering Science. 65, 3279–3292.

Zeng, Q., Guo, Y., Niu, Z. and Lin, W. 2013. The absorption rate of CO_2 by aqueous ammonia in a packed column. Fuel Processing Technology. 108, 76–81.

Zhao, X., Smith, K.H., Simioni, M.A., Tao, W., Kentish, S.E., Fei, W. and Stevens, G.W. 2011. Comparison of several packings for CO_2 chemical absorption in a packed column. International Journal of Greenhouse Gas Control. 5, 1163-1169.