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Biogas Upgrading to Biomethane by CO_2 Removal using Water Absorber with Microbubble Technique

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Abstract

Biogas upgraded to biomethane can be utilized as a renewable energy source to substitute LPG in households and industry. This study explored biogas upgrading by CO_2 removal from 20 - 75 % CO_2-N_2 simulated biogas mixture. The experimental unit using the microbubble technique combined with the water absorption column was set up and used for CO_2 removal from the gas. Microbubble sizes of 20 - 30 µm were generated by a venturi ejector and measured with an automated bubble size measurement. The experiments confirmed that a microbubble with an inline mixer could enhance the effectiveness of the absorption process. The tests demonstrated over 85.80 % removal of CO_2 from the simulated biogas by the experimental unit. The effects of various parameters, including the size of venturi ejector, gas flow rate, water flow rate, liquid-gas ratio, and initial concentration of CO_2, were investigated. The results revealed that 2 L/min gas flow rate, 15 L/min water flow rate, L/G ratio 7.5, and venturi ejector size 0.50 inches are the optimum conditions. The use of the tube absorber gave much higher CH_4 recovery than an absorption column. The appropriate operating conditions gave over 96 % CH_4 concentration or less than 4 % CO_2 concentration, matching the CH_4 purity required by biomethane specifications. The results indicated that the new technique demonstrated in this study can upgrade biogas to biomethane.

Keyword: Biomethane, CO_2 removal, Microbubble, Renewable energy, Water absorption

Introduction

Currently, over 85 % of the global energy demand is supplied by fossil fuels. With increasing concerns of a potential global energy crisis and environmental pollution, the development of clean and renewable energy is a strategic issue connected with environmental protection [1]. Alternative sources of energy like biogas are desired, also as environmentally friendly options. Biogas mainly consists of methane (CH_4, about 50 - 70 %) and carbon dioxide (CO_2, about 30 - 75 %) with some other gases (about 2 %) [2]. High content of CO_2 in biogas decreases its heating value, causes incomplete combustion, unstable flame, and produces a poor performance of the combustion system [3]. Therefore, it is necessary to upgrade the biogas by reducing the proportion of CO_2, to produce biomethane with 90 % or better methane content. Such biomethane can be used in place of LPG in household and industry.

Various techniques can be applied to remove CO_2 from biogas to produce biomethane. Among the most commonly used technologies are adsorption, chemical absorption, pressure swing adsorption (PSA), cryogenic separation, biological methane enrichment, and membrane separation. However, these methods require large investments, have high operating costs, and cause environmental pollution [4]. One interesting method is water absorption that is directly dependent on solubility and contact area between gas and liquid phases. The solubilities of CO_2 and CH_4 in water at 30 °C are 1.25 and 0.0175 g/g H_2O,

respectively, so CO_2 is 70-fold more soluble than CH_4. CO_2 has different electronegativity and becomes surrounded by the polar water molecules, forming a cage structure; this contributes to its comparatively high solubility. A review of CO_2 removal from biogas by water washing is available [5]. CO_2 removal ratio increased from 34.6 to 94.2 % at elevated pressure. [6] investigated water scrubbing for CO_2 and H_2S removal from biogas from manure. The CO_2 removal efficiency was 21.2 % and H_2S removal efficiency was 32.8 %. Microbubble has a large interfacial contact area with the liquid, facilitating absorption of a gas by the liquid. Moreover, microbubble has a low rise velocity from small buoyancy in relation to viscous drag, and correspondingly a long residence time contributing to effective gas absorption [7-10].

There are a few published studies on the application of microbubble. In a water treatment process [11] microbubble with less than 58 μm diameter increased the total mass transfer of ozone and gave higher concentrations of dissolved ozone than a bubble contactor. Moreover, [12] applied microbubble to improve tar removal. A venturi scrubber that can generate microbubble provided an increased absorption surface area and a tar removal efficiency of about 97.7 %.

A water absorption column with microbubble spread throughout the liquid with long contact times seems advantageous. Microbubble of 20 - 30 μm can be generated with a venturi ejector that has high shear forces in its divergent section. The bubbles then tend to gradually decrease in size and may eventually even disappear due to the dissolution of the interior gas into the surrounding water [13]. The use of inline static mixers after the venturi ejector can provide excellent mixing that also increases the mass transfer of CO_2 from gas to liquid phase, to maximize the absorption of CO_2 by the water.

Therefore, this research aimed to study biomethane production by removal of CO_2 from biogas using a water absorption column with microbubble. Venturi ejector with inline static mixer was used to effectively generate microbubble and mix them well in the water flow. A laboratory-scale unit with a maximum gas flow rate of 10 L/min was coupled in a series with 20 - 75 % CO_2 simulated biogas preparation units, a microbubble generator, and water absorption columns. The efficiency of the system was calculated from the contents of CO_2 in entering and exiting gas mixtures.

Materials and methods

Materials

CO_2 and N_2 gases from gas cylinders were used to produce simulated biogas for CO_2 removal testing. The pure CO_2 and N_2 gas cylinders were purchased from Linde (Thailand) Public Company Limited. Tap water without any purification was used as a CO_2 absorbent. Biogas used as feed gas stream in this work was obtained from a wastewater treatment system of Songkla Canning PCL. and had the composition shown in **Table 1**.

Table 1 Composition of biogas.

Compound	Concentration
Methane (CH_4); volume %	69.94
Carbon dioxide (CO_2); volume %	27.25
Hydrogen sulfide (H_2S); ppm	> 800
Oxygen (O_2); volume %	< 0.5
Nitrogen (N_2); volume %	2.2

Experimental

Simulated biogas preparation

The pure CO_2 and N_2 gases from gas cylinders were introduced into and mixed well in a gas mixing tank to produce CO_2-N_2 simulated biogas having 20 - 75 % CO_2. The gas flow rates and concentrations were measured with rotameters and controlled to the desired levels by manually adjusting valves, shown in the diagram of **Figure 1**. Gas samples were taken using a silicone tube, gas sampling pump, and 1 L sampling bags. The gas concentrations were analyzed by gas chromatography with a TCD detector and Porapak Q-packed column.

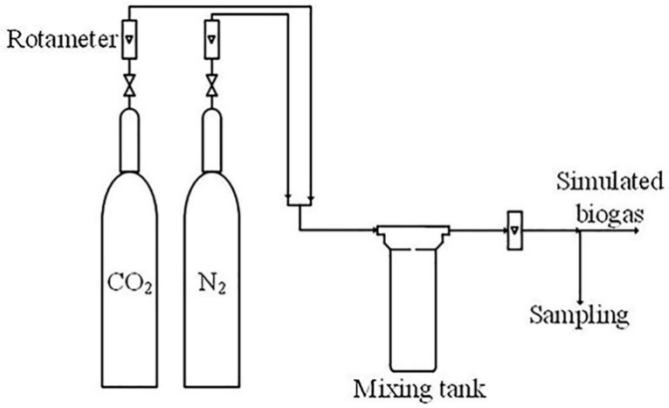

Figure 1 A schematic diagram of the simulated biogas generation system.

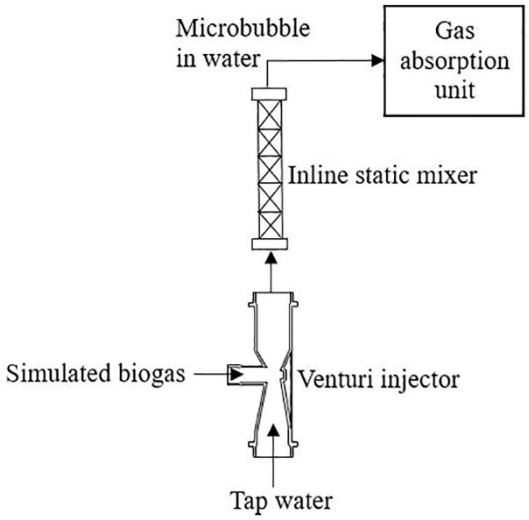

Figure 2 Microbubble generator and inline static mixer.

Microbubble generator

In this research, a venturi ejector was used to generate microbubble. The venturi ejector with its 3 unique sections, including the liquid inlet, the gas suction throat, and the outlet, is shown in **Figure 2**. Tap water was fed in through the liquid inlet and to the constriction area at the throat, where a low-pressure

zone is created, and simulated biogas is sucked in through the suction manifold. The water along with the gas traversed the remaining section of the venturi ejector, where microbubbles were generated by high shear in the diverging part. Gas microbubble was, further, mixed with the water in an inline static mixer. In this manner, the simulated biogas bubbles were efficiently distributed with high contact surface area in the water fed to the CO_2 absorption unit.

Microbubble size determination

Microbubbles are tiny spherical bubbles with a diameter less than or equal to 50 μm [14]. In this study, a microbubble was created to increase the gas-to-liquid interface for mass transfer. This, in turn, should increase the efficiency of CO_2 transfer contact between gas bubbles and the water. Images of a microbubble in water were taken before entering the inline static mixer for size monitoring. Cumulative size distribution of the bubbles was determined by a MATLAB image segmentation program [15]. In a typical microbubble image of 4.2 pixel was the equivalent of a micrometer. Based on the recorded images, the cumulative size distributions were determined. First, the program finds the bubbles that are nearly circular and estimates the radius. Then the outlines of all microbubble that the program has identified are mapped back onto the original image. From the image generated, the sizes of microbubbles can be collected and analyzed in Microsoft Excel.

Absorption column with microbubble generator

The CO_2 removal system was designed and implemented for studying biomethane production and is shown in the diagram of **Figure 3**. The system consists of a simulated biogas generator, gas absorption column, microbubble generator, and 100 L water storage tanks with a water pump.

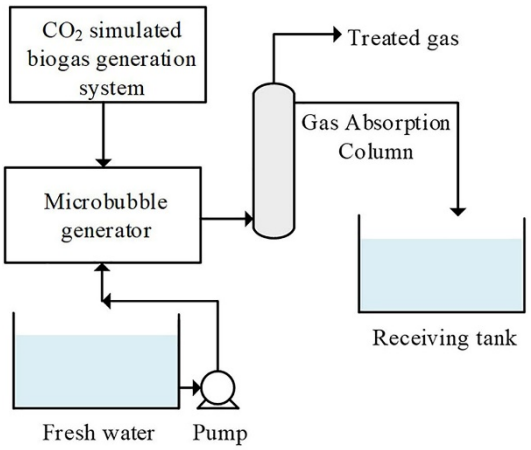

Figure 3 A schematic diagram of the experimental setup for CO_2 absorption in water with microbubble generation.

The 25 L vertical absorption column had 1 m height and 0.18 m diameter. In each experimental run, the simulated biogas and tap water were introduced to the microbubble generator. The gas bubbles in water were transferred to the bottom of the absorption column. The liquid water with microbubble travels through the column while gas is absorbed and dissolved in the liquid phase. The treated gas mainly consisting of N_2 is separated and released at the top of the column.

Tube absorber and gas separation unit

A tube absorber with 10 m length and 0.016 m diameter (ID) was tested in place of the vertical absorption column in the CO_2 removal unit, for possibly increasing the CO_2 removal efficiency. After the

tube absorber, a gas separation column (height 1 m and diameter 0.18 m) was installed to improve the treated gas recovery from water, as seen in **Figure 4**. The water from the tube absorber was continuously fed to the top of the gas separation column and sprayed through a spray nozzle. CO_2-rich water was transferred out and collected in the water receiving tank through the bottom line. The system operation needs at least 10 min to reach a steadystate before taking gas samples.

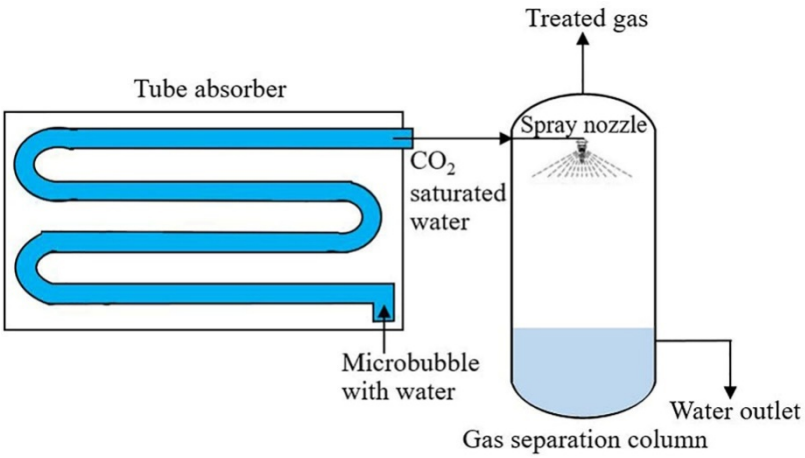

Figure 4 A schematic diagram of the tube absorber with a gas separation column for CO_2 removal.

Experimental operating conditions

To evaluate the performance of the proposed method, sets of experiments were performed with 40 % CO_2 content in the simulated biogas, gas flow rates of 2 - 10 L/min, liquid flow rates of 9 - 17 L/min, and the venturi ejector sizes of 0.25 and 0.50 inch. The concentrations of CO_2 in the simulated biogas entering and exiting the process were determined. The process parameters and experimental conditions are listed in **Table 2**.

Table 2 Experimental operating conditions.

Parameter	Conditions
Size of venturi (inches)	0.25 and 0.50
Gas flow rate (L/min)	2 - 10
Liquid flow rate (L/min)	9 - 17
Initial CO_2 concentration (% CO_2)	20 - 75
Temperature (℃)	25
Pressure (kPa)	101.325

Biogas used as feed gas stream in this testing was derived from the wastewater treatment plant. In an analysis of the biogas composition CH_4 was about 69.94 %, CO_2 about 27.25 %, and H_2S more than 1000 ppm. An H_2S removal unit was installed in this system at the biogas feed line, namely a water spray column with steel wool as packing. The H_2S removal column had a diameter of 0.076 m and a height of 1 m with 0.8 kg of steel wool. Then, the cleaned biogas was transferred to the CO_2 absorption section with a microbubble generation system, to remove CO_2.

Gas sample analysis and calculation

The gas samples were collected in 1 L gas sampling bags. The concentrations of CO_2, CH_4, and N_2 in the entering and treated gas streams were analyzed by gas chromatography. The concentration of H_2S in biogas was analyzed with a gas detector tube. The CO_2 removal efficiency of the system was calculated as follows.

$$\eta_{CO_2} = \frac{[CO_{2,inlet}]-[CO_{2,outlet}]}{[CO_{2,inlet}]} \times 100 \quad (1)$$

Here η_{CO_2} is CO_2 removal efficiency, [$CO_{2,\ inlet}$] is the inlet volumetric flow rate of CO_2 (L/min), and [$CO_{2,\ outlet}$] is outlet volumetric flow rate of CO_2 (L/min).

Results and discussion

Size of microbubble

The size of **a** microbubble in water flow, generated by the 0.50-inch venturi ejector, was determined by using a MATLAB image segmentation program. The microbubble size was measured at simulated biogas flow rates of 2, 4, 6, and 8 L/min with a CO_2 concentration of 40 % at the constant water flow rate of 15 L/min. Bubble images and sizes at various gas flow rates are shown in **Figures 5** and **6**. The results indicate that all gas flow rates gave microbubble in the size range of 20 - 30 µm. The size of a microbubble increased with the gas flow rate. This can be attributed to the growth mechanisms of microbubble and to force analysis on generating the bubbles, such as shear and buoyant forces [16,17]. In [16] the bubbles grew faster at a higher gas flow rate, so their volumes became bigger. In addition, [18] observed that the energy dissipated when bubbles merge leads to the formed larger bubble wobbling. The least size of about 20.83 µm was observed at the gas flow rate of 2 L/min.

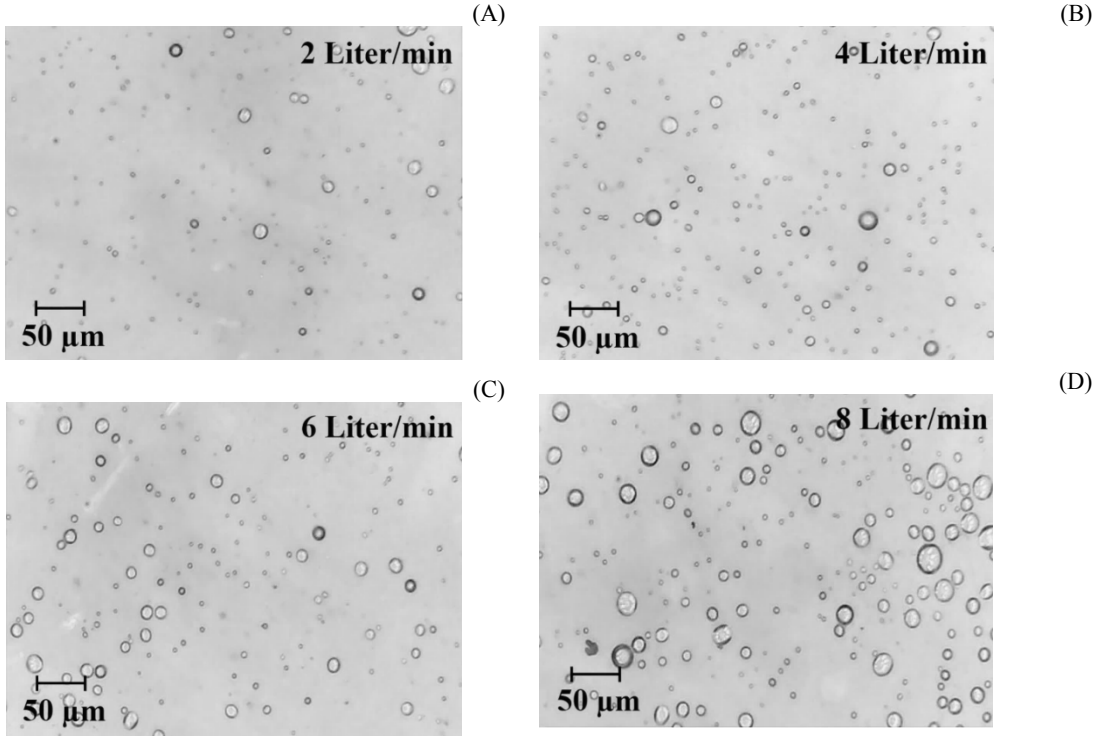

Figure 5 The gas bubbles imaged at gas flow rates of 2 L/min (A), 4 L/min (B), 6 L/min (C), and 8 L/min (D).

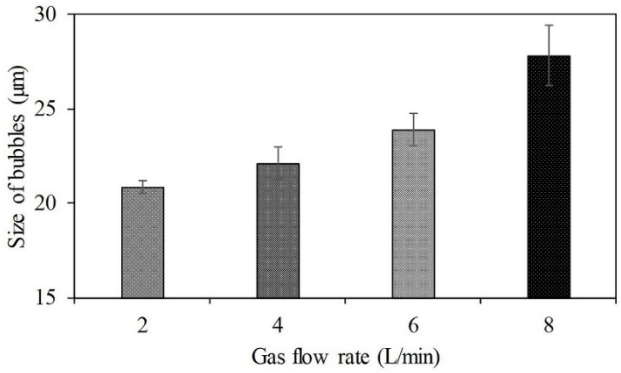

Figure 6 The size of gas bubbles with the various simulated biogas flow rates fed to the venturi ejector.

Effect of venturi ejector size on CO_2 removal

The size of venturi ejector generating microbubble affected CO_2 removal efficiency, as shown graphically in **Figures 7** and **8**. The test conditions were controlled at 2 - 10 L/min gas flow rate, 9 - 17 L/min water flow rate, and 40 % CO_2 in the simulated biogas. We observed that the venturi size of 0.50 inches provided better CO_2 removal at all gas and liquid flow rates, probably because the 0.25-inch venturi had a larger pressure drop affecting the microbubble distribution in liquid water. Therefore, the 0.50-inch venturi ejector was chosen for all following experiments. **Figure 7** also shows the results from increasing simulated biogas flow rate, namely decreased efficiency of the system. This may be because the size of the microbubble increased and gas retention time decreased following the increasing gas flow rate [16,19]. The effect of water flow rate on CO_2 removal efficiency is also presented in **Figure 8**. We observed that both 0.25-inch and 0.50 inch venturis gave similar trends in CO_2 removal, which increased with the water flow rate.

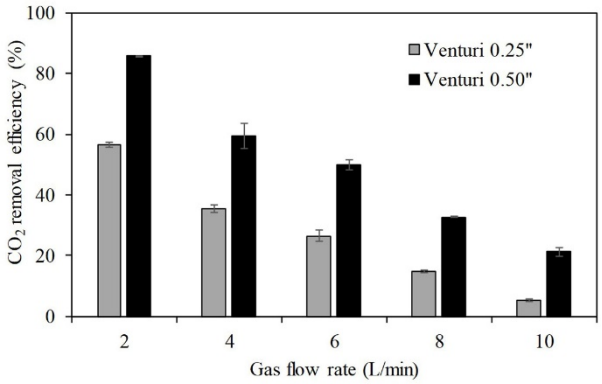

Figure 7 The effect of venturi ejector size at various gas flow rates on CO_2 removal from simulated biogas.

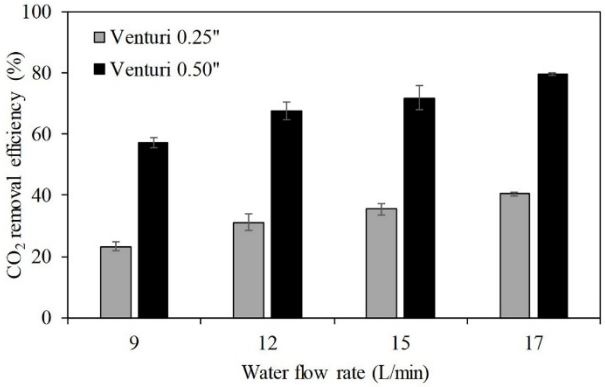

Figure 8 The effect venturi ejector size at various water flow rates on CO_2 removal from simulated biogas.

Effect of gas flow rate on treated gas composition

To study biomethane production, the effects of 2 - 10 L/min simulated biogas flow rates fed to the venturi ejector were observed on CO_2 removal efficiency and treated gas concentration. The water flow rate of 15 L/min and 40 % CO_2 content in simulated biogas were held fixed. The CO_2 can be removed by physical absorption through the microbubble surfaces to form $H_2CO_{3(aq)}$ and $CO_{2(aq)}$ in water, following equation (2). Only a small portion of the aqueous CO_2 is in the form of H_2CO_3 because the equilibrium constant of $H_2CO_{3(aq)}$ is very small (~1.7×10^{-3}).

$$CO_{2(g)} + H_2O_{(l)} \leftrightarrow H_2CO_{3(aq)} + CO_{2(aq)} \quad (2)$$

CO_2 removal efficiency by absorption column

CO_2 removal efficiency versus gas flow rate was calculated and is shown in **Figure 7**. The highest removal efficiency of about 85.80 % was achieved with the lowest simulated biogas flow rate of 2 L/min. The gas flow rate was affected via both absorption capacity and contact time of gas and liquid. Increasing the gas flow rate increased the amount of CO_2 to be absorbed, while the capacity to absorb it was limited. This is due to the shorter gas-liquid contact time with an increased gas flow rate [19]. Hence, the rapid reaction decreased mass transfer and CO_2 removal efficiency. Indeed, on increasing the gas flow rate, the large amount of CO_2 molecules available to diffuse toward the gas-liquid surface, while the availability of liquid is constant, results in limited CO_2 absorption in water [20]. The observed effects of gas flow rate match the prior studies [20,21]. The contacting time or retention time of gas and liquid absorbent can be calculated as follows.

$$T = \frac{V}{Q} \quad (3)$$

Here T is retention time (min), V is absorption column volume (m^3), and Q is outlet volumetric flow rate of CO_2 simulated biogas stream (L/min). It was observed that on increasing the gas flow rate from 2 to 10 L/min, the retention time dropped from 12.50 to 2.50 min. The highest retention time of 12.50 min contributed to CO_2 removal efficiency.

Effect of water flow rate on CO_2 removal

The effects of water flow rate fed to the venturi ejector on CO_2 absorption are depicted in **Figure 8**. The water flow rates of 9, 12, 15, and 17 L/min were tested at the constant simulated biogas flow rate of 4

L/min and the fixed CO_2 concentration of 40 %. The experimental results reveal that increasing the water flow rate could improve CO_2 removal efficiency. The highest 79.60 % removal was observed at the maximum tested 17 L/min water flow rate. This is consistent with the studies [20,22]. A larger amount of water provided increased absorption capacity, so more CO_2 can dissolve in water to form carbonic acid [23,24]. This observation matches [23] which attributed it to the increased bulk absorption capacity of liquid due to there being more liquid.

Effect of liquid-gas ratio

The liquid-gas (L/G) ratio of flow rates was varied by adjusting the gas flow rate to 2, 4, 6, 8, or 10 L/min at the water flow rates of 9 or 17 L/min, as illustrated in **Figure 9**. The CO_2 content in the simulated biogas was held fixed at 40 %. The highest 85.80 % CO_2 removal efficiency was observed with an L/G ratio of 7.5 at a liquid flow rate of 15 L/min. Lowering the liquid flow rate decreased both the L/G ratio and CO_2 removal efficiency. This observation matches the findings in [22,25], which attributed it to the decreased effective interfacial area between liquid and gas phases. Hence, there was poorer availability of liquid molecules to react with CO_2 per unit time with a decreased liquid flow rate. Besides, this result is consistent with the studies [5,21] presenting that a lower L/G ratio results in a relatively faster CO_2 absorption, which makes the mass transfer area decrease.

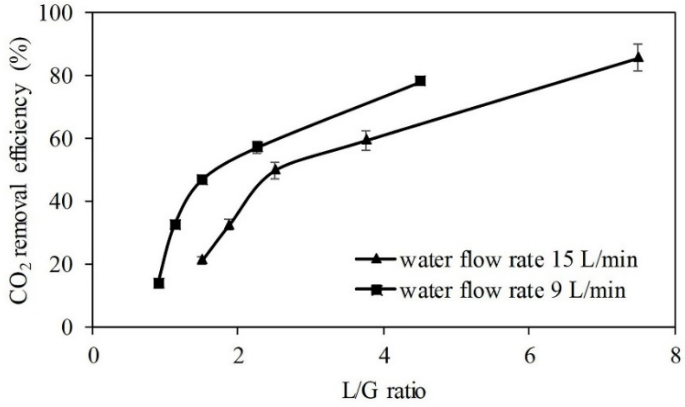

Figure 9 The effect of L/G ratio at different water flow rates on CO_2 removal efficiency in an absorption column with microbubble.

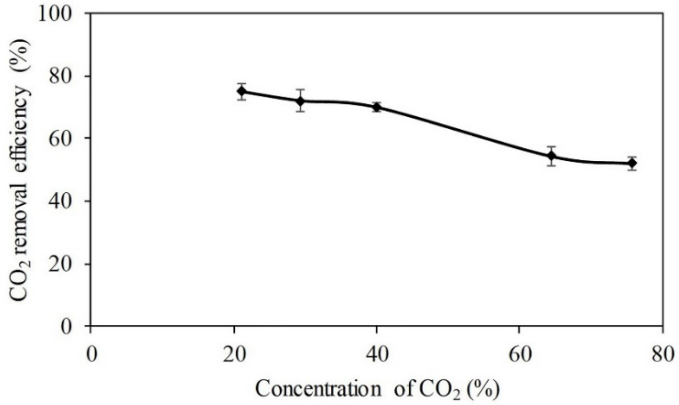

Figure 10 The effect of initial CO_2 concentration in simulated biogas on CO_2 removal efficiency using absorption with microbubble in an experimental unit.

Effect of CO_2 concentration on the absorption

Figure 10 shows the effects on CO_2 removal efficiency of initial CO_2 concentration in the range 20 - 75 % in the simulated biogas. These experiments had a fixed gas flow rate at 4 L/min and a water flow rate at 15 L/min, at ambient pressure and temperature. The CO_2 removal efficiency decreased from 74.90 to 51.90 % as the CO_2 concentration in the simulated biogas increased from 20 to 75 %. This may be because of the limited CO_2 quantity that can be dissolved in the absorbing water flow [2].

CO_2 removal by a tube absorber

Effect of gas flow rate

Figure 11 presents the effects of gas flow rate on CO_2 removal efficiency on using a tube absorber. The experiments were performed by varying the gas flow rate from 2 to 10 L/min and keeping the water flow rate constant at 15 L/min. The CO_2 removal efficiency increased from 42.15 to 91.53 % as the gas flow rate decreased from 10 to 2 L/min. From the plot, it is observed that the tube absorber provided better CO_2 removal efficiency than the absorption column. The head of liquid on the bottom of the absorption column had a significant effect, decreasing the mass transfer coefficient of CO_2 from gas phase to liquid phase [26].

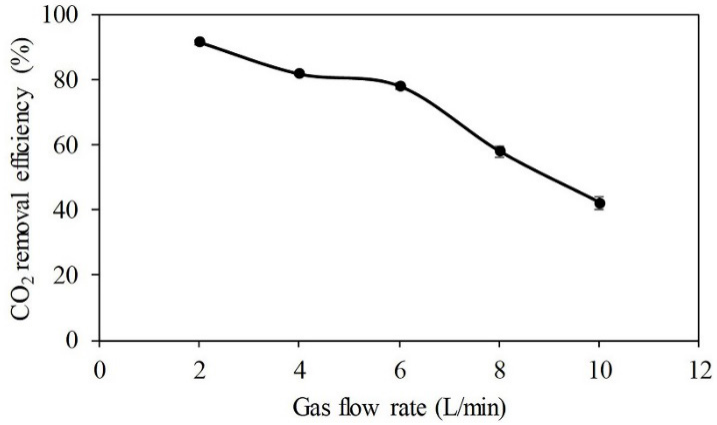

Figure 11 Efficiency of CO_2 removal versus simulated gas flow rate, on using a tube absorber.

Recovery of treated gas

The recovery of treated gas from water-absorbent by a gas separation column was tested at the gas flow rates of 2 - 10 L/min with a water flow rate of 15 L/min. The treated gas recovery of absorption column and tube absorber with gas separation column was investigated. The result showed that with the absorption column an 86 % recovery of treated gas was obtained, while recovery up to 99 % was achieved by using the tube absorber with a gas separation column. It is observed that the gas separation column provides a higher treated gas recovery because spraying the liquid at the top of the column allows effective separation and release of the treated gas from the water-absorbent.

CO_2 removal efficiency in continuous operation

The CO_2 removal efficiency by a continuous process (with water recirculation) is shown in **Figure 12**. 4 L/min gas flow rate, 15 L/min water flow rate, and initial CO_2 concentration at 40 % were held constant. The results at various operating times from 0 to 120 min were recorded. It is observed that the highest CO_2 removal efficiency of about 84.70 % was achieved in the 1st 5 min. The efficiency then decreased with operating time until a very low efficiency of about 37.84 % was reached after 120 min. The CO_2 removal efficiency decreased with time, probably because the water-absorbent became saturated with CO_2 [19].

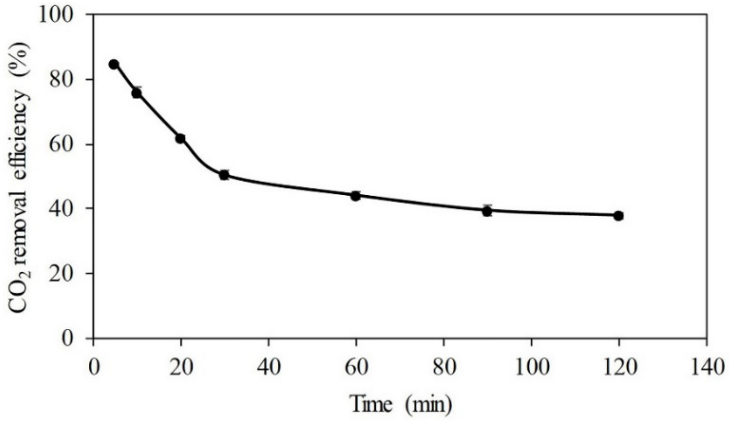

Figure 12 CO_2 removal efficiency versus operating time during a continuous absorption run with microbubble.

Removal of CO_2 from biogas

The biogas from the wastewater treatment system of Songkla Canning PCL. was run through the biomethane production system while using the tube absorber with microbubble generation. The concentration of H_2S in the biogas feed stream was treated to lower than 100 ppm before sending it to the system. The CO_2 removal efficiency was investigated at an appropriate operating point based on the previous experiments, by fixing the 4 L/min of gas flow rate and the 15 L/min of water flow rate. The CO_2 solubility in water is about 70-fold higher than that of CH_4. Therefore, in the absorption process, CH_4 in biogas should remain in the treated gas and not get dissolved in water. Only CO_2 should be removed from the gas blend by absorption. The concentration of CO_2 decreased from 27.25 to 3.27 %, while the CH_4 concentration increased from 69.94 to 96.36 %. The outlet CH_4 concentration was higher than 90 %, satisfying the specifications for biomethane [27]. Also, it was found that the methane loss in the system was only about 0.013 %.

Concerning the CO_2 removal from biogas using water absorption, our results are similar as reported in [5]. The focus of that study was the influences of pressure and temperature. The tests were performed over the pressure range of 0.8 - 1.2 MPa and the temperature range of 7 - 40 °C. In our work were performed at ambient pressure and room temperature using real biogas. Moreover, the tests involved microbubble formation in a venturi ejector which enhanced the absorption capacity. [28] observed CO_2 and H_2S removals using chemical absorption in a packed reactor. The observations revealed a CO_2 removal efficiency of over 90 %, effectively enriching the CH_4 component. However, they also reported that chemical absorption was inappropriate due to requiring a large volume of liquid solvent and having environmental impacts.

Conclusions

The experimental unit for CO_2 removal from CO_2-N_2 simulated biogas was designed for using water absorption with microbubble generation. A venturi ejector generated microbubble of size 20 - 30 µm, with a high interfacial area facilitating CO_2 dissolution into the water. The optimum operating conditions for this experimental setup were 2 L/min gas flow rate, 15 L/min water flow rate, L/G ratio 7.5, and venturi ejector size of 0.50 inches. This study confirmed that the microbubble of the simulated biogas gave over 85.80 % CO_2 removal efficiency by water absorption. Increasing the L/G ratio (increasing water flow rate and decreasing gas flow rate) can improve the CO_2 absorption and give smaller microbubble sizes. With an absorption column, an 86 % recovery was obtained, while recovery up to 99 % was achieved with the use of a tube absorber that provided 96.36 % CH_4 concentration at a suitable

operating point. Therefore, the produced biogas can be used as a renewable fuel in transport vehicles or other power generation.

Acknowledgements

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