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## The Effect of the Packing Flow Area and Biogas Flow Rate on Biogas Purification in Packed Bed Scrubber

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### ABSTRACT

The objective of this study was to reduce the level of impurities in biogas to obtain a higher concentration of methane gas (CH<sub>4</sub>) in it. The biogas purification process was carried out in a packed scrubber using Monoethanolamine (MEA) compound as an absorbent. This research focused on the effect of the packing flow area and the optimum biogas flow rate for obtaining purified biogas with a high concentration of methane (CH<sub>4</sub>). The results of the study reveal that the packing flow area measuring 0.1963 cm<sup>2</sup> is more optimal in the purification process compared to 1.7633 cm<sup>2</sup> packing flow area. Different biogas flow rates at 0.3 L/min, 0.5 L/min, 1 L/min, and 12 L/min yield different results, and the highest concentration of CH<sub>4</sub> at 90.141% is obtained from the slowest flow rate, which is 0.3 L/min. The slow flow rate and a small packing flow area equal to a longer contact time between MEA and the biogas flowing through it; hence, the absorption contact area is also greater compared to that with a faster flow rate; therefore, the highest level of CH<sub>4</sub> is obtained at the slowest biogas flow rate.

Keywords: biogas purification, MEA, packing flow area, biogas flow rate, packed column.

### INTRODUCTION

One of the main matters in facilitating the socio-economic development of a country is energy (Nwokolo et al., 2020). The need for energy has consequently increased along with the increasing world population (Nwokolo et al., 2020). The world's energy reserves are still very much dependent on fossil fuel sources, such as crude oil, coal, and natural gas (Kalsum et al., 2020). However, the use of fossil fuels to fulfill these energy needs leaves a negative impact (Nwokolo et al., 2020). Fossil fuels sources are not renewable and eventually they will be depleted; therefore, renewable energy source is needed (Kalsum et al., 2020). Biogas is one of the renewable energy sources because it uses natural and renewable materials. Biogas has generated massive research interest for production and emerged as one of the alternative fuels (Tetteh et al., 2018). The other advantage of biogas is the waste of biogas named slurry can also

be converted into organic fertilizer. Biogas is a flammable gas that mainly consists of methane  $(CH_{4})$  and carbon dioxide  $(CO_{2})$  and is obtained from organic compounds decomposition by anaerobic bacteria (Speight and Radovanovic, 2020). The biogas compositions commonly consists of 50-70% methane, 30-40% carbon dioxide, and other gases in small amounts, and the composition is depending on the type of the organic compounds from (Iswanto et al., 2021). Biogas has a heat combustion value between 4800 and 6200 kCal/m<sup>3</sup>, with a specific gravity that is 20% lighter than air (Mara, 2012). Biogas has been widely used in the community, including in biogas stoves and lamps (Abdurrakhman et al., 2020). Biogas with methane content will generate a fairly clean combustion without producing soot (Kasikamphaiboon et al., 2013). The combustion value of pure methane gas can go as high as 8900 kCal/m<sup>3</sup> (Mara, 2012). However, methane is also one of the greenhouse gases with a negative impact that is 21 times more harmful

than  $CO_2$  (Gustiar et al., 2014). Nonetheless, the negative impact of methane can be reduced by using it as fuel (Allo and Widjasena, 2019). Besides its advantages, there is also a drawback from the unpurified biogas, which comes from the impurities within it, such as carbon dioxide  $(CO_2)$ and hydrogen sulfide (H<sub>2</sub>S). The CO<sub>2</sub> and H<sub>2</sub>S content in biogas will lower its calorific value and trigger corrosion respectively, which the impacts lead to decreasing quality of biogas (Tabatabaei and Ghanavati, 2018; Seohartanto et al., 2021). Moreover, H<sub>2</sub>S and CO<sub>2</sub> in biogas can also pollute the environment and are detrimental to human health (Saleh et al., 2015a). To reduce the impact of gas impurities, biogas has to be purified before use. After undergoing a purification process, biogas can be used as co-generator fuel (Detman et al., 2017). In addition to its use as fuel for household needs, the biogas with a high enough methane content can also be used as fuel for CNG machines in industry (Saleh et al., 2015).

One of the methods of biogas purification is absorption. The absorption method is widely used to remove the CO<sub>2</sub> and H<sub>2</sub>S content in the chemical industry (Tabatabaei and Ghanavati, 2018). In the absorption process, there is a substrate that absorbs particular substrates depending on the substrate solubility, it is named absorbent. The absorption process in biogas purification is called gas absorption, it occurs when the biogas is in contact with the absorbent solution, and the absorbent absorbs the impurity gases. In principle, the separation is based on the solubility of the diluted (impurities) component in the absorbent; and methane will not be dissolved in the process (Singhal et al., 2017). Therefore, it is necessary to choose a selective absorbent that does not absorb methane gas. The absorption method is designed to operate in biogas low residence time at ambient pressure and temperature (Tabatabaei and Ghanavati, 2018).

The tool used in the study is a packed scrubber, and the packing type used is raschig ring type with two kinds of flow areas. A packed scrubber is basically a column filled with filler material (Ardhiany, 2018). In this matter, the filler refers to packing. A scrubber is a tool used to capture and remove unwanted substances (Setyowati, 2017). The objective of this study is to increase the CH<sub>4</sub> concentration using monoethanolamine (MEA) as an absorbent in the packed scrubber, while taking into account the different packing flow areas and biogas flow rates.

### MATERIALS AND METHODS

#### Experimental set up

Prior to the process of purification of biogas in a packed bed scrubber, several steps must be conducted which are the preparation of material slurry of cow dung and biogas production. The main tools to produce biogas and to purify biogas have been integrated into one unit in order to make the process of anaerobic digestion and gas purification easy to be controlled.

In the preparation process, cow dung which is used as feed in biodigester is mixed with water on a scale of 1:3 to form slurry. Then, the mixture is mixed properly and added a 50 ml prebiotic in order to increase biogas production. The type of biodigester that is used is the fixed dome with a total volume of 250 liters. Moreover, the slurry is added to the biodigester until it is filled 80% of the total volume of biodigester, other 20% of the volume of the biodigester is left empty to make a space where biogas will be formed.

Biogas purification is carried out during the optimum time of biogas production, i.e., on the 21st day of biogas fermentation, Tetteh Emanuel et.al.,2018 stated that optimum time of anaerobic digestion is on the 20th day. Biogas purification is carried out using the absorption method (Daiyan et al., 2020) by allowing it to pass through the scrubber, which will absorb the impurities such as H<sub>2</sub>S and CO<sub>2</sub> by absorbent solution. The shape of Packed Bed Scrubber for purification process is a cylindrical column, in addition of support plate inside in order to support packing materials. This is used in the process of purification. Well designed packed bed scrubber will provide the required mass transfer contact between gas and liquid phase and will give high removal efficiencies of impurities. Below is the Figure 1 of Packed Bed Scrubber that is used in this study.

Raw biogas which is produced by biodigester is pumped from bottom column to the upper part of the scrubber column; meanwhile, the MEA 1 M solution is pumped from the upper part of the scrubber column to bottom column. The MEA solution contacts the biogas stream trough the packing. In the study, a variety of biogas flow rates rate were used, i.e. 0.3 L/min, 0.5 L/min, 1 L/min, and 12 L/min. As for the packing, we are using the raschig ring type with different flow areas, i.e, 1.7663 cm<sup>2</sup> and 0.1963 cm<sup>2</sup>. The formula for calculating the packing flow area (a) is as follows:  $a = \frac{\pi D^2}{4} (\text{Kern, 1983}) \tag{1}$ 

where:  $a - \text{flow area (cm}^2)$ , D - diameter packing (cm).

In the Figure 2 below are the types of packing used in the study of the Packed Bed Scrubber. The results of the purification process were analyzed using gas chromatography (GC) to find out methane concentration in the gas, whereas the  $H_2S$  and  $CO_2$  concentration was measured using biogas analyzer device. The research process is briefly illustrated in Figure 3 below.

### **RESULTS AND DISCUSSION**

### Biogas composition before purification

The research was carried out on the 21st day of biogas production. On that day, the concentration of methane gas was quite high and there were no signs of abating; therefore, it was safe to surmise that the concentration of carbon dioxide was also high; hence, the gas could be purified by reducing the  $CO_2$  concentration in the raw biogas. The data pertaining to the analysis of biogas composition before purification are presented in Table 1 below. The results of methane chromatogram analysis from the gas chromatography (GC) are presented in Figure 4 below.

# The effect of the packing flow area (a) on methane gas $(CH_4)$ concentration in purified biogas

Packing flow area is the extent of an area that can be passed through by both the absorbent and gas during the biogas purification process using



Figure 1. Packed bed scrubber

the absorption method. The effect of the flow area on the yield of biogas purification is presented in Figure 5 below. Figure 5 shows that a flow area measuring 1.7663 cm<sup>2</sup> will yield 80.484% of



Figure 2. Raschig ring packing, (a) 1.7663 cm<sup>2</sup> flow area, (b) 0.1963 cm<sup>2</sup> flow area



Figure 3. Research process schematic for absorption process

 Table 1. Data before biogas purification

Composition	Concentration	
CH <sub>4</sub> (%)	60.231%	
CO <sub>2</sub> (%)	26.60%	
H <sub>2</sub> S (ppm)	263 ppm	

purified biogas; meanwhile, a 0.1963 cm<sup>2</sup> flow area will yield 90.141% of purified biogas, which is an increase from the figure before purification at 60.231%. To express it in another way, the size of a packing flow area definitely affects the yield of the purification process. i.e. a smaller flow area will yield more purified biogas. This is because a smaller flow area will allow for a longer contact time, and therefore it will be able to absorb more impurities. The findings are in line with a study by Kadarjono et al., (2020) which show that pall ring is the most effective packing among

pall rings, ralu rings, and nor-pac rings because it creates a smaller dimension. Nonetheless, care must be taken to ensure that the diameter or a flow area is not too small because it may obstruct the flow of the fluid. The result of this study is consistent with the previous study by Kadarjono et al. (2020) that states the effective packing area tends to diminish, as the fluid flow rate increases. This is also in line with the results of a previous study by Arachchige and Melaaen (2012), which compared the type and size of packing that the use of random packing with a smaller packing size will capture more CO<sub>2</sub>, while the use of structured packing will capture more CO<sub>2</sub> if the packing size is larger. The volumetric mass transfer coefficient commonly increases along with surface area (Kolev et al., 2006). However, the packing surface area should not be the only scoring criterion for estimating a higher mass transfer



Figure 4. Methane composition before purification



Figure 5. The effect of the flow area and biogas flow rate on the yield of purified biogas

coefficient; there are also other influencing factors such as packing arrangement pattern, crimp height, and crimp angle in the case of structured packing (Aroonwilas et al., 2003).

## The effect of the biogas flow rate on methane gas $(CH_{4})$

The effect of the biogas flow rate on the rising concentration of methane gas (CH<sub>4</sub>) is presented in Figure 5. Other gases such as CO<sub>2</sub> and H<sub>2</sub>S have greater solubility compared to CH<sub>4</sub>. Moreover, Figure 5 also shows that the concentration of methane gas increased to 70-90% in relation to the size of the packing flow area. Before purification, the concentration of methane in biogas was 60.231%, after the purification process with 0.3 L/ min flow rate using 1.7663 cm<sup>2</sup> packing flow area, the concentration rose to 87.484% and the figure rose even further to 90.141% with a packing flow area measuring 0.1963 cm<sup>2</sup>. The graph shows that a faster flow rate will lower the yield of methane gas. A fast flow rate means a shorter contact time between the gas and MEA in the packed scrubber; hence, fewer impurities will be absorbed by MEA. It also mentioned by Villadsen et al., (2021) observed that the increase in gas flow decreased the percentage of removal. The findings of this study are in line with the findings of the previous study by Kasikamphaiboon et al., (2013), which reveals that the slowest biogas flow rate at 1 L/min will yield the best results in gas impurities absorption.

## The effect of the biogas flow rate on carbon dioxide (CO<sub>2</sub>) concentration

Carbon dioxide  $(CO_2)$  is the by-product of a perfect combustion process; hence,  $CO_2$  is a substance that does not burn again. Therefore, when

 $CO_2$  is present in fuel, it will reduce the calorific value of the fuel. Biogas contains a significant amount of  $CO_2$  (ranging from 25–45%). Reducing the  $CO_2$  concentration in biogas will significantly increase its quality; therefore, it must be done. The  $CO_2$  absorption reaction is presented below (Kasikamphaiboon et al., 2013).

$$CO_2 + 2RNH_2 RNHCOO^- + RNH_3^+$$
 (2)

The analysis of the CO<sub>2</sub> concentration after biogas purification reveals a significant drop. After purification, the level of CO<sub>2</sub> can be lowered from 26.60% to 0.20–2.75% depending on the different biogas flow rate being used. Meanwhile, the effect of the biogas flow rate on the composition of carbon dioxide (CO<sub>2</sub>) is presented in Figure 6 below.

Figure 6 above shows that excellent absorption occurs when the biogas flow rate is at 0.3 L/min. After the CO<sub>2</sub> purification, the concentration dropped to 0.34% with 1.7663 cm<sup>2</sup> packing flow area and dropped even further to 0.20% with 0.1963 cm<sup>2</sup> packing flow area. MEA itself is a relatively strong alkali with a fast reaction rate and is able to lower the CO<sub>2</sub> concentration (Dang and Rochelle, 2003). There is a reversible and exothermic reaction between MEA and CO<sub>2</sub> by supplying heat to the system (Krumdieck and Wallace, 2008).

From the figure it can also be learned that the faster biogas flow rate would lead to a higher  $CO_2$  concentration after purification. A fast flow rate means a shorter contact time between biogas and MEA in the packed scrubber; hence, less impurities will be absorbed by MEA, whereas a slow flow rate will lead to more  $CO_2$  being absorbed, because there will be more contact time and more contact area between the biogas and MEA, which will lower the concentration of  $CO_2$  after purification and increase the concentration of methane in biogas. This is due to gas solubility, wherein  $CH_4$ 



Figure 6. The relationship between carbon dioxide (CO<sub>2</sub>) and the biogas flow rate

Table 2. H<sub>2</sub>S concentration before and after purification

Packing flow area (cm <sup>2</sup> )	Biogas flow rate (L/min)	H <sub>2</sub> S before purification (ppm)	H <sub>2</sub> S after purification (ppm)
	0.3		0
1.7663	0.5	263	0
	1.0		0
	12		0
	0.3		0
0.1963	0.5	263	0
	1.0		0
	12		0

has a lower solubility than  $CO_2$ . However, unlike the results obtained by Augeletti et al., (2020), the higher the biogas flow rate, the greater the total amount of  $CO_2$  absorbed. There are differences in the research techniques, where Augeletti et al., (2020) compares three variations of biogas flow rate with the same absorption time, whereas this study was not at the same absorption time but by filling the sample bag to the brim with the same size sample bag.

## The effect of the biogas flow rate on hydrogen sulfide (H<sub>2</sub>S) concentration

Hydrogen sulfide  $(H_2S)$  is a colorless gas and its presence in biogas will cause an unpleasant odor, and in high concentration it will affect respiratory health. For that reason, the  $H_2S$  concentration in biogas has to be lowered or removed entirely. Biogas purification using MEA with different packing flow areas and different biogas flow rates shows that the  $H_2S$  concentration can be dropped from 263 ppm to 0 ppm. The effect of the biogas flow rate on the composition of hydrogen sulfide ( $H_2S$ ) is presented in Table 2. The results of the analysis reveal that the  $H_2S$  concentration dropped from 263 ppm to 0 ppm after undergoing the purification process with MEA 1 M solution with different biogas flow rates. H<sub>2</sub>S has been identified as the substance that causes problems in equipment due to its corrosive nature that will corrode and form crust in equipment. Using biogas while it still contains H<sub>2</sub>S will produce sulfur and sulfuric acid that are corrosive to any types of metal. Removing H<sub>2</sub>S from biogas through the purification process will protect the machine components from the corrosive effect of biogas. Even at a concentration as low as 0.0005 to 0.3 ppm humans can detect the smell of H<sub>2</sub>S. At a higher concentration, H<sub>2</sub>S may cause a person to lose his/ her sense of smell (Sianipar, 2009). The biogas purified with MEA solution will have all traces of H<sub>2</sub>S completely removed, as indicated by the loss of the typical foul odor of hydrogen sulfide  $(H_2S)$ .

### CONCLUSIONS

The results of the study show that the best biogas flow rate in the purification process that will yield the highest  $CH_4$  concentration at 90.141% is 0.3 L/min, which is the slowest flow rate with a packing flow area measuring  $0.1963 \text{ cm}^2$ . The best combination of the biogas flow rate and packing flow area size at 0.3 L/min and 0.1963 cm<sup>2</sup> will lower the concentration of CO<sub>2</sub> and H<sub>2</sub>S to 0.20% and 0 ppm. This is because a slow flow rate will lead to a longer contact time between biogas and MEA and the extent of the contact area; therefore, more impurities will be absorbed and in turn will lead to a higher yield of CH<sub>4</sub>.

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