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Analysis of Flow Rate and Concentration of Mono Ethanol Amine (MEA) in the Biogas Purification Using Absorption Column with Pall Ring Packing

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Abstract*— The usage of energy derived from fossil sources can contribute to increasing greenhouse gas emissions such as carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). One of the alternative energy sources with high potential for development in Indonesia is biogas. Animal waste can be processed into biogas using anaerobic digestion technology. This research aims to develop science in the use of biogas from cow dung as a renewable energy source. The study focuses on analyzing the flow rate and concentration of Mono Ethanol Amine (MEA) in the absorption column. The first stage in this research is to produce biogas from cow dung using a fixed dome type biodigester with a COD content of 33,438 mg/L, TSS of 25,000 mg/L, a C/N ratio of 16.1, and a pH of 6.7. Fermentation in the biodigester is carried out for 30 days, and the biogas produced is analyzed every 5 days. At the highest methane content of biogas yield, the next stage is to purify the biogas from impurities using an absorption column. This absorber uses pall ring-type packings made of plastic with dimensions 1 x 1,5 cm. The biogas flow rate into the column is 0.5 L/min, and the absorbent solution flow rate is varied at 0.5, 1, and 1.5 L/min, and the MEA concentration is varied at 1 M, 3M, 5M, and 7 M. The result showed that the optimal flow rate of the absorbent solution was 0.5 L/min, and the optimal concentration of the absorbent solution was 1 M. These conditions resulted in the CH_4 content of methane gas increasing from 55.27% to 85.84%. The findings of this study support the use of absorbent MEA in a packing column absorber for purifying biogas from cow dung.

***Keywords*— Cow dung; biogas purification; Mono Ethanol Amine (MEA); absorption column.**

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I. INTRODUCTION

Energy has been a vital and central driver of a country's socioeconomic development. As the world population continues to grow, energy demand is expected to rise accordingly. The energy used by the world's population is currently dominated by fossil energy. According to [1], fossil fuels have been utilized at a high rate as the primary energy source for industrial processes and daily use. The result is an increasing crisis of global energy and environmental problems, as well as a rise in the rate of carbon dioxide (CO2) emissions. In Indonesia, demand for fuel oil and natural gas (LPG and LNG) is predicted to increase. An increase in energy sources does not accompany the need for this increase; an increase in the use of alternative energy is highly expected.

The use of energy derived from fossil sources contributes to the rise in greenhouse gas emissions, including carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). Greenhouse gas emissions primarily originated from specific CO2-emitting sectors within the industry, including coal-fired power plants, the petroleum industry, the cement industry, and the steel mill industry [2]. Several technologies are employed to reduce CO2 emissions, including absorption, adsorption, membrane separation, freezing technology, and water scrubbing [3], [4]. Energy from renewable sources must be continuously developed, especially from biomass. Sources of biomass energy include crops, wood residues, municipal waste, and animal waste [5].

One of the alternative energy sources with high potential for development in Indonesia is biogas. Biogas has low carbon content, so it does not cause emissions; the carbon produced from biogas ranges from 50 to 450 g CO2eq/kWh [6]. Animal waste, municipal waste, agricultural and industrial waste, such as sewage sludge, crop waste, food processing residues, and wet animal manure, can be processed into biogas using anaerobic digestion technology [7], [8], [9]. Anaerobic digestion is defined as a microbiological process in

the decomposition of specific organic matter, commonly in the absence of oxygen. This concept has been widely accepted and is easily found not only in many natural environments but also widely applied in today's technology for producing biogas through an air-tight reactor tank, commonly known as digesters. This process involves an extensive range of microorganisms, resulting in two main end products: biogas and digestate. Biogas usage is primarily associated with electricity and heat generation, as well as household cooking and heating [10]. Biogas can be defined essentially as combustible gas, while digestate can be best described as a decomposed substrate consisting of rich macro- and micronutrients. Those substances are beneficial; for example, they can also be used as a plant fertilizer. Anaerobic digestion presents an opportunity for resource recovery, waste minimization, and GHG emission reduction. Through anaerobic digestion, organic matter in manure is broken down by microbes in an oxygen-deficient environment, producing methane-rich biogas [7]. Anaerobic digestion is a natural biological degradation process based on a series of biochemical and physical-chemical reactions performed by a complex ecosystem of microorganisms. This ecosystem in an anaerobic environment gradually converts organic matter into a biogas enriched in methane and a biofertilizer [11]

The main components of Biogas include methane (CH4), carbon dioxide (CO2), and trace amounts of hydrogen sulfide (H2S), however, this also varies depending on the source of biogas [12]. The source of organic waste and production pathways influence the composition of biogas [13], [14]. Between these gaseous compounds, CO2 and H2S are acid gas impurities that can potentially decrease the quality of the biogas and become a major factor in corrosion problems of the process equipment. This will potentially delay the rate of chemical combustion reactions [15]. Biogas as an energy source utilizes its methane gas content (CH4), while in the production of biogas, methane gas is still mixed with other gases such as CO2 and H2S, and a small amount of other gas content [16]. An appropriate processing strategy is required to condition several process parameters for fermentation, including pH, temperature, C/N ratio, fermentation time, and others. This adjustment is necessary when performing a methane gas purification process to achieve a high concentration of methane gas. Conditional to the nature of the substrate and the pH of the reactor, biogas can produce a range of 50–70% CH4 and CO2 at a concentration of 30–50%, with additional minor components including hydrogen sulfide (H2S), nitrogen (N2), oxygen (O2), siloxanes, volatile organic compounds (VOCs), carbon monoxide (CO), and ammonia (NH3) [1].

Various purification methods for biogas include physical absorption, chemical absorption, and membrane separation. The absorption method is a popular method to remove CO2 and H2S in the chemical industry [13], [17]. In the research conducted by [18] using membrane technology with 30% MEA as an absorbent, CO_2 absorption reached 96%. The CO2 and H2S content in biogas should be considered since these also affect the yield and quality of the biogas to be used. The CH4 content in biogas can be increased by 75–98% while reducing the carbon dioxide (CO2) and hydrogen sulfide (H2S) content. The high H2S content in biogas causes corrosion of metal components within the engine. H2S is an inorganic acid that attacks the surfaces of a metal when placed in direct contact. Sulfur stress cracking (SSC) is a widely known corrosive mechanism that occurs when a metal meets H2S. This process occurs when the H2S concentration is above 50 ppm [19]. Carbon dioxide reacts with the amine, which increases the solubility [19]. One of the processes of separating CO_2 from a gas stream is by the absorption method [20]. In this process, CO_2 contained in the gas stream is absorbed into the liquid solution due to the concentration difference, and the CO2 absorption process is limited by mass transfer. To increase mass transfer, the contact area between the gas phase and the liquid phase must be efficient [21].

Absorption columns have several types, such as bubble absorption columns, plate absorption columns, packing absorption columns, and spray absorption columns [15]. The selection of the type of packing is done by considering the pressure drop and flooding aspects. Therefore, in this study, selecting a packing material with small dimensions and good performance will be a priority. The types of packing that are often used in industry, such as Raschig rings, Pall rings, Tellerette, Intalox/Metal, Bert Saddle, and Intalox/Saddle [22].

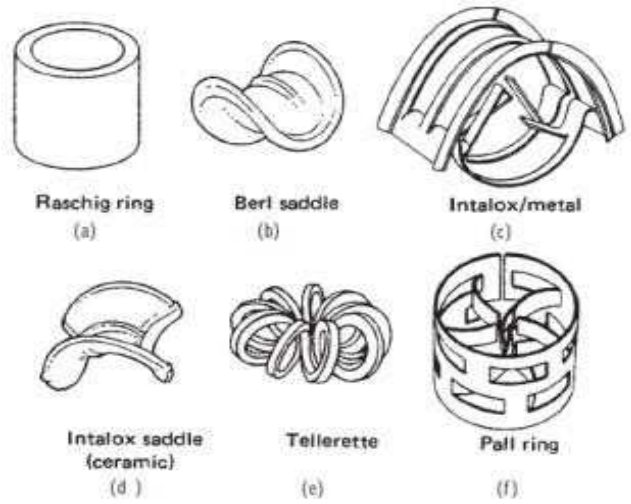

Fig. 1 Types of Packing [22]

In the industrial world generally, the chemicals used for the absorption process are classified as primary amines (such as Monoethanolamine/MEA) [23], secondary amines (such as Diisopropanolamine/DIPA), tertiary amines (such as triethanolamine, TEA), and steric amine (such as 2-amino- 2-methyl-1-propanol, AMP) [14]. The amine process constitutes over 60% of existing CO_2 capture units [24], [3], [25].

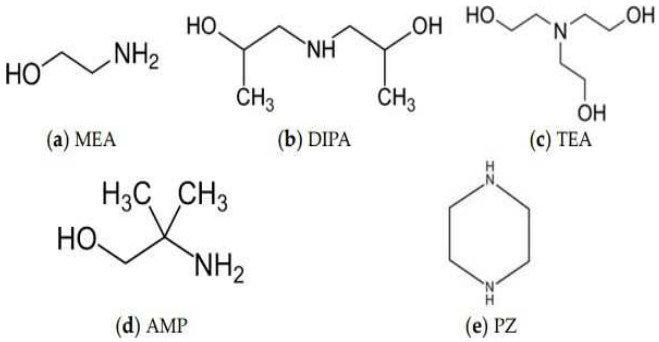

Fig. 2 Structure of Various Amines [2]

TABLE I
PHYSICAL AND CHEMICAL PROPERTIES OF AMINES

Items	MEA	DIPA	TEA	AMP	PZ
M.W (g/mol)	61.084	133.19	149.188	89.138	86.136
Density (g/cm3)	1.0117	0.992	1.126	0.934	1100
BP (C)	170	249	335.4	165.5	146
Solubility	Miscible	Miscible	Miscible	Miscible	Miscible
Vapor Pressure (pa)	64	2	1	40	10.66
pKa	950	8.80	7.76	9.70	9.78
Reaction rate constant	3630	2585	2202	810.4	48.533
Activation energy Ea (kj/mol)	41.2	39.9	36.9	41.7	33.7

The mixtures of Alkanolamines have been developed, MEA remains the most appropriate choice for the absorption of CO2 and H2S due to its high reactivity. MEA is also cheap, has a low molecular weight, and is stable over temperature [24], [26]. Using a suitable solvent is needed due to the impact of eliminating corrosion, and the need for a tremendous amount of energy to regenerate the solvent, as well as the possibility of absorbing more CO2 content. MEA includes a primary amine compound containing one amine group and can absorb CO_2 and H_2S. The equation that describes the reaction between MEA with H_2S and CO_2 in the absorption process is as follows:

$2RNH_2 + CO_2 \longrightarrow RNH_3^+ + RNHCOO^-RNH_3 + CO_2 + H_2O \longrightarrow RNH_4^+ + HCO_3^- RNH_2 + H_2S \longleftrightarrow RNH_3^+ + HS^-$

AMP is an amine that has a high absorption capacity and low energy consumption for regeneration, but its reactivity to CO_2 is lower compared to MEA. While PZ has a high CO_2 absorption capacity, the disadvantage is that PZ can only precipitate at high concentrations. To improve the performance of amine solvents, mixed amines are formulated to combine the advantages of each amine, and another amine can compensate for the disadvantages of one amine. Research [27] shows that TEA is the best amine in CO_2 absorption in their research, with the addition of HCO_3 as a single absorption product, HCO_3 is more effective in the hydrogenation process compared to carbamate added to a mixture of MEA and DEA amine. Based on research conducted by [27] it was indicated that the amine mixture (MEA+AMP) is the best compound for O_2 absorption. Meanwhile [21] In his research, he used a mixture of amine AMP+PZ+MEA with the following sequential concentration ratios (1,5M - 1,5 M – 3M), and the research results showed that the mixture was effective for use as an alternative absorber, as indicated by the increase in CO_2 efficiency.

II. MATERIALS AND METHOD

The focus of this research is to determine the optimal flow rate and concentration of MEA absorbent in the biogas purification process. The primary equipment used in this research was a biodigester and an absorption column, along with supporting tools including a Nova Plus gas analyzer, COD analyzer, TSS analyzer, C&N analyzer, pH meter, and pressure gauge. The process stage carried out in this research is preparing cow dung slurry, carrying out the anaerobic fermentation process, and purifying the biogas produced from the biodigester. In preparing cow dung slurry by mixing cow dung with water in a 1:3 ratio, the slurry is analyzed for its characteristic COD, TSS, C/N, and pH.

The process of fermenting cow dung in a biodigester involves feeding slurry into the biodigester, filling it to 80% of its total volume of 250 liters, and then continuing with fermentation for 30 days. Based on the results [23] of previous research on biogas fermentation from cow dung, the highest methane gas concentration was achieved on the 21st day by 60,23%. On the day the highest biogas concentration is achieved in this research, biogas samples are taken for purification. Measuring the biogas concentration is done by first flowing the biogas produced from the biodigester into a gas bag. Then the gas flows into the Nova Plus gas analyzer to measure its concentration.

The purification process in the absorption column is carried out by flowing biogas into the column in the opposite direction to the MEA absorbent flow. The absorption column uses plastic pall ring-type packing with dimensions of 1 x 1,5 cm. The choice of pall ring column in this study is because the pall ring has a good balance between surface area and vacuum so that it will improve mass transfer performance [13] [19]. In addition, the low-pressure drop in the pall-ring column will facilitate higher gas production and stable pall-ring operating conditions at various flow rates, allowing for operation using different solvents. The MEA solutions used were 1M, 3M, 5M, and 7 M. The Biogas flow rate was 0,5 L/min. and the MEA flow rate was varied to 0.5 l/min, 1 l/min, and 1.5 l/min. The determination of MEA and biogas flow rate is based on previously conducted tests to assess the equipment's capabilities. The purified biogas is then analyzed for its CH_4, CO_2, and O_2 content using a biogas analyzer.

This research follows the research flow diagram, as shown in Figure 3. The leading research equipment consists of a biodigester and an absorption column, as illustrated in Figure 4.

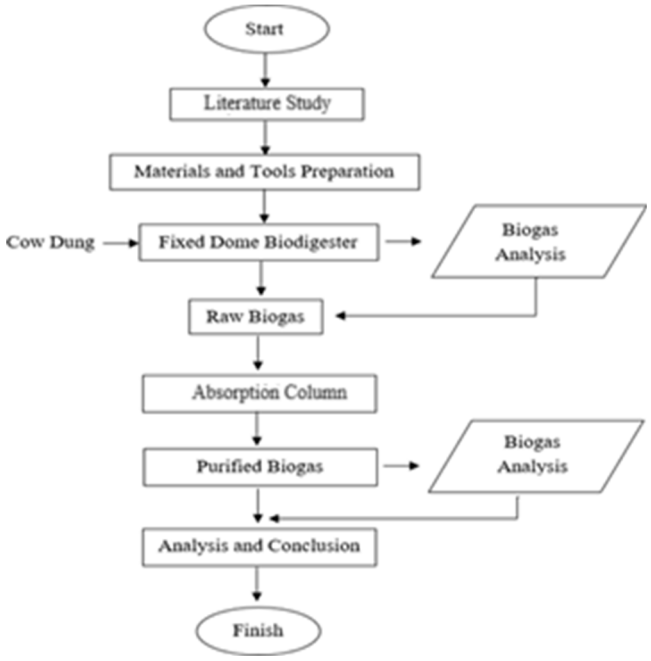

Fig. 3 Research Flow Diagram

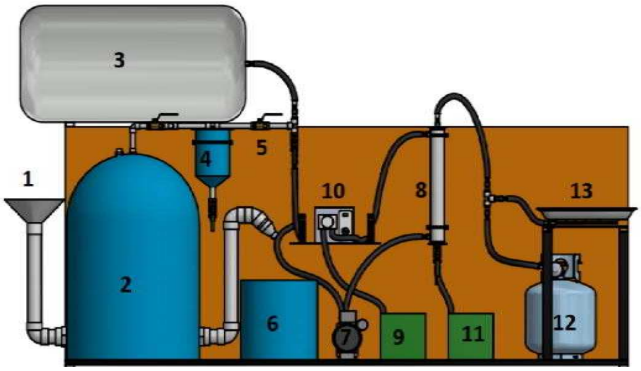

Fig. 4 Biogas Unit

Notes:

1. Feed input
2. Digester
3. Biogas Storage
4. MEA Storage
5. Valve
6. Sludge Storage
7. Compressor
8. Absorption Column
9. MEA lean
10. Pump
11. MEA rich
12. Biogas Tank
13. Stove

III. Results and Discussion

Cow dung, as a raw material for biogas, has been analyzed before the fermentation process in the biodigester. The analysis aims to determine the characteristics of cow dung slurry, including COD, TSS, pH, and C/N ratio. From Table 2, which is the result of cow dung analysis, it can be stated that the organic content COD of 33.438 mg/l and TSS of 25.432 mg/l of cow dung is considered as high enough to be processed into biogas, and the pH of 6.7 was also sufficient to perform the fermentation process.

TABLE II
DATA FROM ANALYSIS OF COW DUNG RAW MATERIALS

Parameter	Value	Unit
COD	33,438	Mg/L
TSS	25,432	Mg/L
C/N	16.15	%
pH	6.7	-

The optimum fermentation process of cow dung in the biodigester was achieved on the 25th day, with a methane content of 55.27% in the biogas. The biogas produced from the digester then continued for a purification process in the absorption column. Based on the research result [28]. The purification of biogas from POME using K2CO3 absorbent solution in an absorption column with packing can reduce the CO_2 content by 47,38%.

TABLE III
BIOGAS FERMENTATION RESULTS FROM THE DIGESTER

Composition	Concentration
CH4 (%)	55.27
CO2 (%)	33.09
H2S (ppm)	476

After performing the purification process with some research variables, the biogas samples were sent to the Sriwijaya State Polytechnic Chemical Engineering laboratory to be analyzed using a multi-detector gas analyzer. The results of biogas purification are presented in Table 4.

TABLE IV
DATA ON BIOGAS PURIFICATION RESULT

MEA Absorbent		Biogas Composition (%)		
MEA Flow Rate (L/min)	MEA concentration (M)	CH_4	CO2	O2
0.5	1	85.84	0	8.83
1		82.33	0.7	11.52
1.5		83.33	0.32	14.08
0.5	3	85.32	0.52	13.33
1		84.80	0.45	13.54
1.5		81.23	0.43	12.55
0.5	5	85.30	0.43	14.3
1		84.41	0.4	14.51
1.5		84.06	0.41	14.21
0.5	7	84.84	0.3	13.33
1		84.37	0.37	13.70
1.5		84.43	0.38	12.0

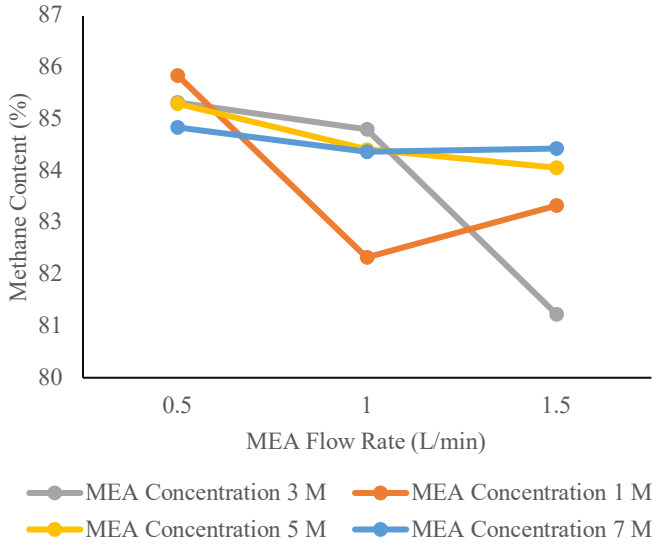

Fig. 5 The effect of flow rate on the CH_4 produced

The MEA flow rate greatly influences the concentration of CH_4 gas produced from biogas after it is purified through an absorbent column. Figure 5 demonstrated that when the MEA flow rate was at its lowest level of 0.5 L/min, a significant increase in CH_4 content was obtained. Before performing purification, the methane content in the biogas was 55.27%. After conducting the purification process at a flow rate of 0.5 L/min using a plastic Raschig-ring packing type and an MEA concentration of 1 M, the methane content increased to 85.84%. Based on the results of the first year's research using a plastic Raschig-ring packing type, it was able to produce methane purity from biogas up to 90.14% from the raw biogas with a methane content of 60% [23].

The graph shows that a faster flow rate will reduce the yield of methane gas. The effect of the resulting flow rate is in line with previous research [23] which also showed that a slower biogas flow rate results in a higher concentration of methane reaching. Fast flow rates result in shorter contact times between the gas and MEA in the absorption column. Hence, fewer impurities will be absorbed by the MEA. This is by the research from [3] where the lowest MEA concentration is the optimal concentration, this is beneficial for the physical absorption of CO_2. For that reason, if the CO_2 concentration decreases, the CH_4 content will increase.

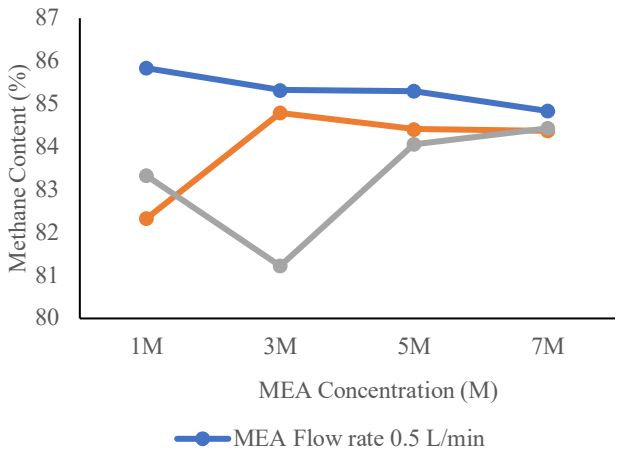

Fig. 6 The effect of MEA flow rate on the CH_4 produced

Variations in MEA concentration will affect the amount of CO_2 absorbed, which will have an impact on increasing of CH_4 gas. According to [14], [23] using MEA can absorb higher levels of CO_2 since chemical reactions occur between primary amines and CO_2 to form carbamates according to [29]. Biogas purified with the MEA solution will eliminate the H_2S impurity gas content, characterized by the disappearance of the characteristic foul odor associated with hydrogen sulfide (H_2S). In biogas purification using MEA with a concentration of 7M, it results in a decrease in CO_2 levels and an increase in methane production to 84.84%.

Seen from Figure 6. Increasing the concentration above 0.5 M does not significantly increase the purity of methane from biogas. This is by the research from [3] where the lowest MEA concentration is the optimal concentration, this is beneficial for the physical absorption of CO_2. For that reason, if the CO_2 concentration decreases, the CH_4 content will increase. In research using water scrubbing with a pressure of 6 to 10 bar, drying of biomethane after the absorption process is required because the methane gas still contains water vapor; in this way, the methane gas content reaches more than 90% [17], This is caused by the fact that CO_2 is easily soluble in water [30]. Thus, in this study, it was found that a 1 M MEA concentration is the optimal concentration for biogas purification, yielding a methane purity of 85.84%. From this research, a reduction of CO_2 was achieved through several variations of MEA concentrations and flow rates, reaching 98%. This is aligned with the research from [16] where MEA is effective in absorbing polluting gases in biogas and capable of removing CO_2 in biogas, resulting in more than 85% methane production. At MEA concentrations of 3M, 5M, and 7M, there is no noticeable increase in methane purity in biogas. This is due to the MEA being only able to achieve its maximum absorption of 85.84% methane purity.

IV. Conclusion

The MEA flow rate significantly affects the concentration of CH4 gas produced from biogas after purification through an absorbent column. Fast flow rates result in shorter contact times between the MEA and gas in the absorption column. Utilizing MEA can absorb higher levels of CO_2 due to the chemical reactions that occur between primary amines and CO_2 to form carbamates. The optimum flow rate and MEA concentration for producing methane with the highest methane content are at a MEA flow rate of 0.5 L/min and an MEA concentration of 1 M, resulting in an increase in methane content from 55.27% to 85.84%.

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