

Quality Improvement of Biogas Fuel through the Adsorption Method

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Abstract

Biogas is an alternative energy that has been developed at this time as a substitute for petroleum energy sources. In producing the biogas, purity is essential such as free from carbon dioxide, hydrogen sulfide, and water vapor. The objective of this study was to develop a biogas purification process that can be used in rural areas using cheap raw materials including dop, knee, T, PVC pipe, aluminum clamps and multiplex boards with techniques that can be operated easily. A method for biogas preparation employed in this study was the adsorption method using solid adsorbents. The adsorption method use zeolite to remove carbon dioxide, iron powder to remove hydrogen sulfide, and silica gel to remove water vapor. The results of biogas purification showed that there was an increase in methane of $18.85 \pm 5.26\%$, a decrease in carbon dioxide of $46.28 \pm 6.05\%$, a decrease in hydrogen sulfide of 100%, effectiveness of water vapor absorption of 36.787 ± 8.515 g water per cubic meter of biogas, and an increase in calorific value of 9.775 ± 0.806 %. Consumption of biogas to ignite lantern lamp and electric lamp bulbs was lower after purification, accounting for 25.93%, and 26.17%, respectively. The cost of removing impurities was estimated at 0.13 USD per cubic meter.

Keywords: Adsorption; Biogas; Fuel; Purification

1. Introduction

Biogas is a renewable fuel gas produced by anaerobic fermentation from organic matter with the help of the bacterium *Methanobacterium sp.* Biogas is a fuel used for cooking and an electric energy generator. The biogas components consist of 50 – 75 % methane (CH_4), 25 – 45 % carbon dioxide (CO_2), 0 – 0.3 % carbon monoxide (CO), 1 – 5 % Nitrogen (N_2), 0 – 3 % hydrogen (H_2), 0.1 – 0.5 % Hydrogen Sulfide (H_2S) 0.1 – 0.5 %, Oxygen (O_2), and water vapor (Wellinger *et al.*, 2009). The only component that functions as fuel is CH_4 . The gas content of H_2S , CO_2 and water vapor are impurities that can not be burned (Hakim *et al.*, 2018)

Biogas purification needs to be an essential consideration because it affects the heat produced so that the biogas produced needs to be purified to increase the purity of CH_4 by removing impurities. The impurities that affect the heat value are CO_2 , water vapor, and H_2S . Water vapor can cause corrosion of the equipment or machinery used (Mamun *et al.*, 2017). The presence of the CO_2 in biogas is highly undesirable, because the higher CO_2 levels in biogas, it will further decrease the biogas heat value and is very disturbing in the combustion process. The presence of H_2S gas interferes with combustion since H_2S gas can transform to sulfur dioxide (SO_2) and sulfur trioxide (SO_3), which are corrosive to equipment such as gas generator when

further oxidized by water vapor to form acids that can damage the equipment components (corrosive). Condensation can be a problem in gas compressors. Biogas purification can remove impurities, thus preventing corrosion of the equipment used.

In biogas purification, there are various technologies developed that are physicochemically and biologically processed. The physicochemical processes include chemical absorption, chemical adsorption, a solid adsorbent, and chemical scrubbing. Biological processes include the biological oxidation in the condition under microaerobic, microbiological with a separate biofilter using *Acidithiobacillus thiooxidans*, bioscrubber, and biofiltration (Axelsson *et al.*, 2012).

In this study, the adsorption technique was applied to purify the biogas because the cost is relatively low and easy to obtain raw materials. The novelty of this research was the use of biogas purifiers which could be accepted by the community, especially in rural areas. The purifier is easy to operate and can significantly increase blue percentage, mitigate odor problem, and prevent damage of gas generator equipment due to the corrosive gases.

Adsorption is the process by which molecules of substances from a gas mixture or liquid solution became attached to a solid or liquid surface. This occurs because of an imbalance of molecular forces in solids, which tend to attract other molecules that adhere to the surface. Materials used as absorbents are generally solid materials. Solid adsorbents that have the potential to purify biogas; for example, are zeolite, silica gel, iron oxide (Fe_2O_3).

The objective of this study is to develop a system that can remove impurities in biogas which uses cheap raw material and can be easily applied in rural areas. Purification of biogas was performed by a adsorption technique in order to minimize CO_2 , H_2S , and water vapor content. This can be also applied for combustion that can speed the cooking process, because the produce heat was larger with less odor and corrosion on the machine components. The use of purified biogas can also reduce greenhouse gas emissions because biogas combustion releases less

nitrogen oxide, hydrocarbons, and carbon monoxide than burning gasoline and diesel (Zhao *et al.*, 2010).

2. Materials and methods

The experiments performed in this study were conducted at the Research Center for Appropriate Technology, Indonesian Institute of Science. The detailed methods were clarified in the sections below.

2.1. Biogas digester

Fixed dome digester used in this study was made of fiberglass with 2.2 meters in height, 1.8 meters in diameter, and capacity of 5.5 m^3 . The fixed dome was the most popular model in Indonesia, where the installation of the digester was embedded in the ground. This allows the conservation of space, the stability of digester temperature, and growth support of methane-producing bacteria.

The digester was equipped with a PVC pipe inlet for the introduction of biogas raw materials and a PVC pipe outlet for discharging the fermentation of residual biogas. The inlet dimension was 31 mm in diameter and 19 mm in height spliced to PVC pipe of 16.5 mm in diameter, 30 mm in height, while the dimension of outlet was 4" in diameter and 22 mm in length. The outlet was operated based on the principles of hydrostatic pressure equilibrium.

2.2. Raw materials for filling biogas digesters

The digester was filled up by cow dung and water was added. The cow dung was originated from cattle breeding of Cikole sub-district, Subang district. The water was added to the cow dung at a ratio of 1: 1 to obtain a dry weight of about 9%. Filling cow dung was performed once per day with a mixture of 100 kg of cow dung and 100 kg of water.

2.3. Measurement of Biogas Production

The biogas produced was measured by a gas meter in specifications as follows: Q_{max} 6 m^3/h , Q_{min} 40 dm^3/h , P_{max} 50 kPa, V 0.7 dm^3 . While the gas meter used to measure the use of biogas for biogas stove and gas generator

were as follows: Q_{\max} 3 m³/h, Q_{\min} 16 dm³/h, temperature -20°C + 50°C, P_{\max} 1.5 bar, V 1.2 dm³. The measurement of biogas production was carried out once per day.

2.4. Biogas purification

Figure 1 shows the schematic diagram of a biogas application for lighting using the biogas purification without and with adsorption technique. In the adsorption technique, the gas from the digester flowed through the purifier column equipped with the gas meter then to the gasholder. The biogas purification method applied adsorption technique for the gas treatment because it is economically attractive, easy to operate, and highly effective in removal

of gas impurities. Adsorption is a process that occurs when a liquid or gas is bonded to a solid and ultimately forms a film layer (thin film) on the surface of the solid (Wahono *et al.*, 2010). The surface adsorption process of solids involved the transfer of solutes in the gas to the solid surface, where the transfer process is driven by the Van der Waals force. Adsorbents used are usually granular in shape having large surface area per unit volume (Yentekakis *et al.*, 2017). In this study, silica gel was used to absorb water vapor in biogas. In contrast, the iron powder (Fe₂O₃) with the a spiral and long shape and paramagnetic properties was produced from the metal casting of metal workshops and used to bind H₂S. Zeolite was also used to remove CO₂ and increase CH₄ purity in biogas.

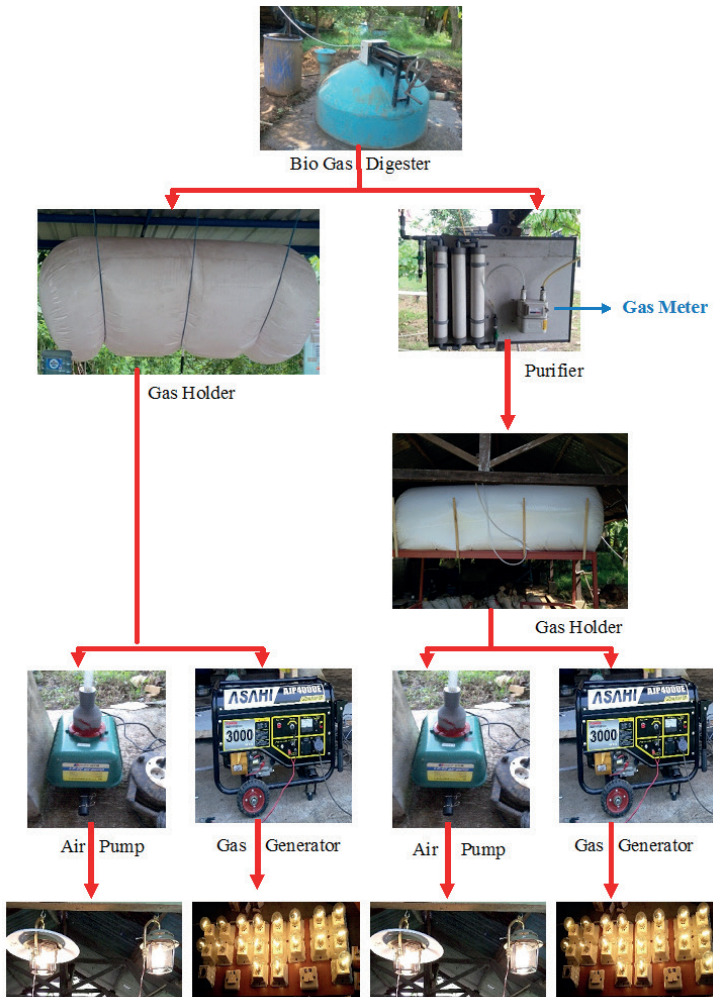


Figure 1. The schematic diagram of a biogas application for lighting

2.5. Analysis of the chemical compound in biogas

Analyses of the chemicals in the original and purified biogas were carried out using a portable biogas analyzer VT-A50x 300 B model, with the range as follows: 0 - 100% v/v CH₄, 0 - 100% v/v CO₂, 0 - 2,000 ppm H₂S, 0 - 25 % v/v O₂, 0 - 10,000% v/v CO, and 0 - 5,000 ppm NO_x.

2.6. Utilization of biogas for lighting

Biogas produced by the digester before and after refining was applied to ignite lantern lamps and electric lamp bulbs. Tests to ignite on lantern lamp were carried out using an air pump to increase biogas pressure with the following specifications: LP 60 mode, 220 V / 240 V input, 50 - 60 Hz frequency, 70 liters/minute output, power of 60 watts, pressure of 0.04 mPa. Tests were performed in duplicates using two lantern lamps. Testing to ignite the electric lamp bulb was carried out at 1000 watts, 1100 watts, 1200 watts, 1300 watts, 1400 watts, 1500 watts, 1600 watts, 1700 watts, and 1800 watts, previously converted to electricity using a gas generator with the following specifications AJP 4000 E type, 220 V rate voltage, 50 Hz rate frequency, 2.5 KVA output rate, 3 KVA maximum output, 3000-watt power generator.

Result and Discussion

3.1. CH₄ content

Biogas contained about 50 - 60% methane which is a fuel, and 40% of the rest were impurities such as CO₂, H₂S, and water vapor. The CH₄ gas levels before and after purification are shown in Figure 2. The impurity gases could not burn and reduce the biogas calorific value and cause corrosion of the equipment used, so that purification is necessary. The presence of such gas in methane was highly undesirable. CO₂, H₂S and water vapor in biogas decreased CH₄ calorific value and interfere with the combustion process, H₂S and water vapor could cause corrosion of machine components, allowing a maximum concentration of 5 ppm. If the biogas is burned, then H₂S will be more dangerous, because it will form new compounds together with oxygen to SO₂ or SO₃ at the same time it will form H₂SO₄ and a more corrosive compound.

The quantity of CH₄ contained in biogas was determined the quality of combustion process. The content of CH₄ at 40 - 50% have been able to become a fuel for cooking, but if it will be used as gasoline, the CH₄ content is at least 70% (Yentekakis *et al.*, 2017). To improve the quality of biogas, purification has to be carried out. CH₄ levels after purification increased by 10.93% (A), 20.15% (B), 15.93% (C), 25.38% (D), 23.35% (E) and 17.35% (F) at an average value of 18.85% ± 5.26%.

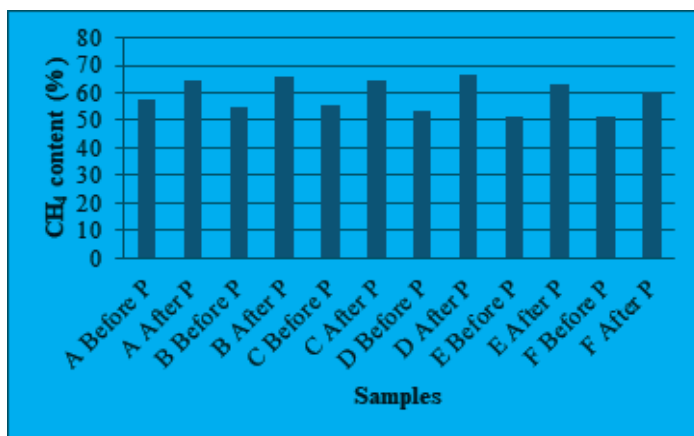


Figure 2. CH₄ gas content before and after biogas purification (P)

3.2. CO₂ content

The increase of CH₄ content was caused by the decrease of CO₂ and H₂S gas contained in biogas as shown in Figures 3 to 5. This was in accordance with the use of zeolite filter Wahono *et al.*, (2010), CH₄ content can increase by 5 - 20% from the initial CH₄ level. Whereas the results of Harihastuti and Sari (2011) research on biogas purification using a multilevel absorption method of activated charcoal absorbent can increase CH₄ by 17.16%. According to Surono and Machmud (2012), CO₂ adsorption process with zeolite adsorber can improve the purity of biogas.

Figure 3 showed the CO₂ gas levels before and after the purification. The use of biogas purification equipment of zeolite material resulted in a decrease in CO₂ content of 43.20% (A), 52.18% (B), 39.81% (C), 39.69% (D), 50.70% (E) and 52.09% (F) at an average value of 46.28% ± 6.0477%. The content of CO₂ before passing purification was 24.35% (A), 24.8% (B), 17.91% (C), 25.7% (D), 21.3% (E) and 26.3% (F) while that after purification was 13.83% (A), 11.86% (B), 10.78% (C), 15.5% (D), 10.5% (E) and 12.6% (F).

To remove 46.28% CO₂, 642.4 g of zeolite was used so that every gram of zeolite can remove 0.072% CO₂. This result was better than the results of Sugiarto *et al.*, (2013) which shows that 150 g of zeolite can remove CO₂ by 29.8% or every gram of zeolite can remove 0.039%

CO₂. The experimental results of Supriyanti (2016) showed that the decrease of CO₂ content with Ca(OH)₂ solution medium reached 30%. The occurrence of CO₂ absorption by zeolites was pointed out from the color of more turbid or duller zeolite when compared to that before purification. Similar results also occurred in a study conducted by Sugiarto *et al.*, (2013) where the zeolite color after refining tended to be darker. In this refining process, CO₂ gas absorbed in zeolite occupied the porous cavity in the zeolite, so that there was no reaction of zeolite evaporation which affected the change of biogas flame to blue, whereas before refining the color of the flame was slightly yellowish.

The polarity of the adsorbed particles caused the affinity of zeolite. Molecules that have a significant permanent quadrupole moment interact strongly with an electric field gradient created by the cation movement of the zeolite (Widhiyanuriyawan *et al.*, 2014).

The absorption of CO₂ by zeolites is according to the polarity that CO₂ has. Because of O atoms with negative charges tend to donate their electron loads to the positively charged zeolite cations on the zeolite surface namely Li⁺, Na⁺, K⁺, Ca²⁺, Sr²⁺, Ba²⁺, Cu²⁺, Zn²⁺, and Mg²⁺. While the cations in the zeolite were first drawn by the oxygen charge on the structure of SiO₄ or AlO₄, the C atoms of CO₂ become unstable, because the O atom donates the electrons, so C tends to be pulled and binds to one of the O atoms on the structure of SiO₄ or AlO₄ (Bonenfant *et al.*, 2008).

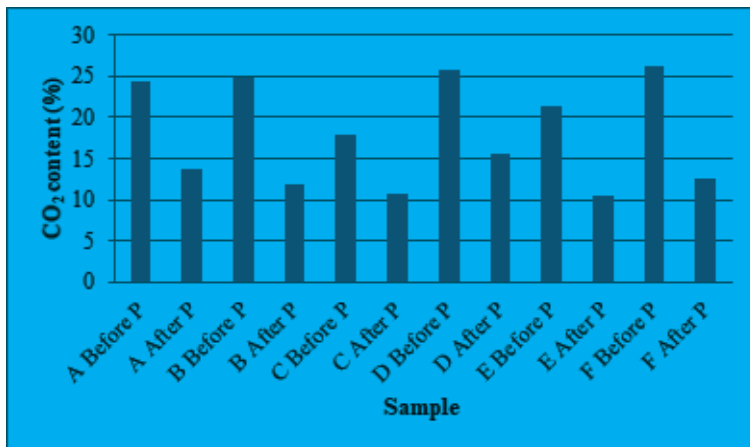


Figure 3. CO₂ content before and after biogas purification (P)

3.3. O₂ content

Figure 4 shows the increase of O₂ content due to the decrease of CO₂. O₂ content increased by 50.43% in A (from 5.81% before purification to 8.74% after purification), 55.96% in B Zfrom 5.7% to 8.89%), 72.77% in C (from 3.82% to 6.6%), 51.02% in D (from 4.9% to 7.4%), 50.943% in E (from 5.3% to 8%), and 52.27% in F (from 4.4% to 6.7%). The O₂ content was increased in the average of 55.57% ± 8.665%. The zeolite weight in the biogas purification device decreased 2.6055 g before purification, and 1.9631 g after purification. The average decrease of zeolite weight was 0.6424 grams or 24.65%.

Increasing O₂ gas content in biogas was caused by the decomposition process of CO₂ gas absorbed by zeolite at the time of the adsorption process to CO and O atom. The absorbed CO₂ gas will be decomposed into one atom C and two O atoms. Atom C will remain trapped in zeolite cavities while O atoms will continue to cause O₂ content to increase. There are three causes of CO₂ decomposition such as thermal reactions, electrolysis, and catalysts (Widhiyanuriyawan *et al.*, 2014). Zeolite has the property of being a catalyst so that the decomposition process of CO₂ can occur. Chemical reactions of CO₂ decomposition is as follow: $2\text{CO}_2 \rightarrow \text{CO} + \text{O}_2$.

3.4. H₂S content

H₂S gas had the following characteristics: colorless but had a distinctive odor such as rotten eggs at low concentrations; often referred to rotten egg gas, is a toxic gas type, flammable and explosive at 4.3% LEL (Lower Explosive Limit) with a blue flame at a temperature of 500°F (260°C). H₂S gas density is heavier than that of air, so taht H₂S gas will tend to be accumulated in a low place. H₂S gas density is about 20% heavier than air with a ratio of specific gravity H₂S to Air is 1.2. H₂S can be soluble, water solubility is 437 ml/100 ml water at 0 °C, or 186 ml/100 ml water at 40 °C. H₂S is corrosive and can cause rust on metal equipment (Negara *et al.*, 2012). Hydrogen sulfide is a weak acid which have $K_a = 1.3 \times 10^{-7}$ mol/L; $pK_a = 6.89$. H₂S content after purification using iron powder decreased to 0 ppm in every experiments from 3124 ppm of A, 2899 ppm of B, 4277 ppm of C, 4019 ppm of D, 3967 ppm of E and 3562 ppm of F, as shown in figure 5. In all measurements, the H₂S reduction was 100%. This was due to the absorption seen in the color of iron powder to black, which indicated the occurrence of H₂S absorption by iron powder. The use of iron powder in the purifier was 737 g, which can eliminate 100% H₂S, every gram of iron powder can eliminate 0.13% H₂S.

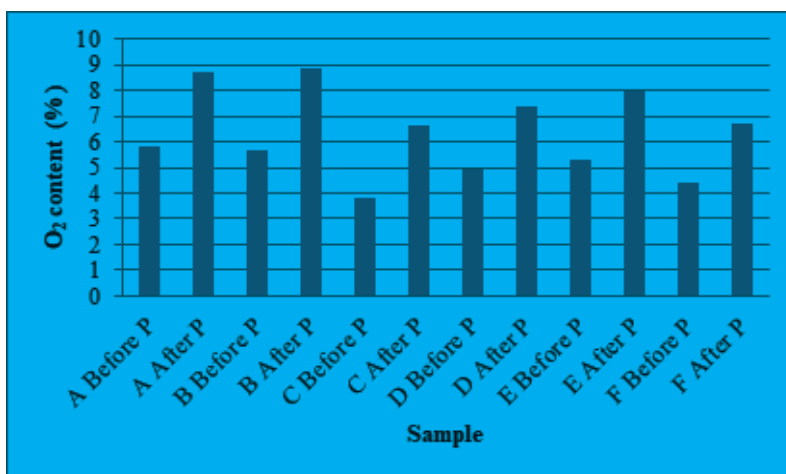
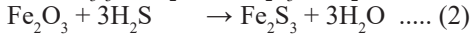
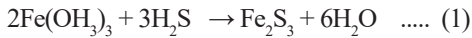


Figure 4. O₂ content before and after biogas purification (P)

The results of this study were higher than that of Negara *et al.*, (2012), which shows that to remove 54% of H₂S 500 grams of iron powder is needed, so every gram of iron powder can remove 0.11 % H₂S.

Van Der Waals force results in the absorption of H₂S content in biogas. The adsorbent can attract the existing H₂S content in biogas. The attractiveness of adsorbent was stronger than the attractiveness of CH₄ (Padang, 2016). The absorption of H₂S in iron powder visually was the change of color of iron powder to black, and caused the smell of biogas is lost.

H₂S will be absorbed into Fe(OH)₃ or absorbed into Fe₂O₃. Using this dry process H₂S is converted to iron (III) hydroxide or iron (III) oxide and water, based on reaction 1 and 2 as follows;



3.5. Calorific value

Figure 5 shows the calorific values of biogas before and after passing through the purifiers. Heat value (HV) is the amount of energy released when a fuel is completely burned in a steady flow process, and the product is returned to the state of the reactant. The calorific value of biogas after purification increased by 8.76% (A), 9.98% (B), 11.12% (C), 9.38% (D), 9.39% (E) and 10,02% (F), with an average of 9.775% ± 0.806%.

This increase in calorific value was caused by the absorption of CO₂ and H₂S gas. The calorific value of biogas burning was 4,800 - 6,900 kcal/m³ (Alayi *et al.*, 2016). According to Surono and Machmud (2012) the electrical power produced by a generator with fuel passing through purification was greater than that produced by a generator with the fuel directly flowing from the digester

3.6. Water vapor absorption

Absorption of water vapor in the biogas was due to the condensate formed can accumulate in the gas channel and can also develop a corrosive acid solution when H₂S is dissolved in water (Wellinger *et al.*, 2009). Figure 6 showed the absorption of water vapor on silica gel adsorbent. The weight of silica gel before purification was determined as 2000 g, 2059 g, 2058 g, 1994 g, 2089 g, and 2121 g for A, B, C, D, E, and F respectively, while after purification it was determined as 2612 g, 4175 g, 4402 g, 4082 g, 4219 g and 4321 g for A, B, C, D, E, F respectively. The efficiency of water vapor absorption for A, B, C, D, E, and F was calculated as 32.66, 27.35, 50.35, 38.85, 30.005, and 41.505 g H₂O/m³ (the average of water vapor absorption efficiency of 36.79 ± 8.151 g H₂O/m³ biogas). Each gram of silica gel absorbs 0.017% gram H₂O/m³ of water vapor. The results of tests conducted

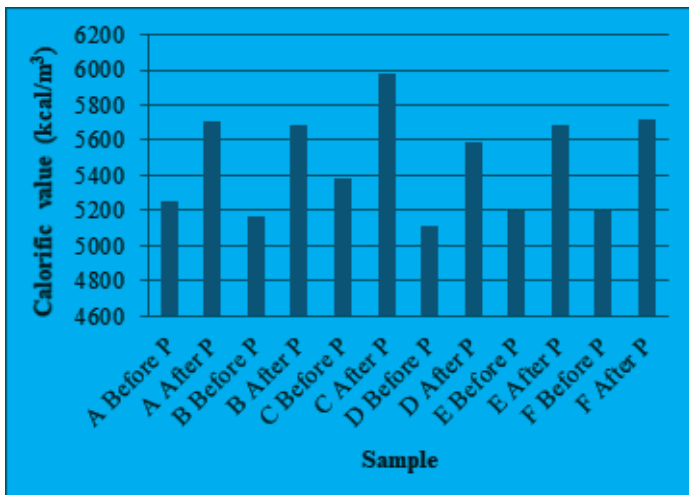


Figure 5. Calorific value before and after biogas purification (P)

by the Energy Conversion Laboratory of Mechanical Engineering of Sebelas Maret University in Surakarta showed that the average effectiveness of H₂O absorption by silica gel was about 4.1 g of H₂O/m³ biogas (Suyitno *et al.*, 2010). Reduction of water vapor is necessary because the condensate formed can accumulate in the gas line and can also create a corrosive acid solution when H₂O is dissolved in water. The longer the purification time, the higher the water vapor absorption. The absorption of water vapor in silica gel was indicated by the color change from blue, before purification, to black after purification.

The use of silica gel adsorbents in water absorption is more effective than that of active carbon. On the use of activated carbon effectivity of water vapor absorption of A and B was 5.39 and 9.45 g H₂O/m³ biogas respectively. The average water vapor absorption was 7.42 g H₂O/m³ biogas.

3.7 Biogas purification costs

The results of research from Wahyu *et al.*, (2012) concluded that the economical minimum capacity for biogas purification is 100 tons/day, with biogas production of 7500 Nm³/day. In this study, the biogas digester used was 5.5 m³ which was a pilot-scale for energy needs for 1 family consisting of 4 people. Stuffed cow dung of 100 kg per day with a biogas production of 3 m³/day or 90

m³/month. Biogas consumption for lighting using 1 lantern 538 L/hr, 2 lanterns 874 L/hr, after biogas purification consumption of biogas for 1 lantern 394 L/hr, 2 lanterns 655 L/hr, can save biogas consumption was around 25.94%. Biogas consumption for lighting using a light bulb was an average of 646 L/hr, after biogas purification consumption of 477 L/hr, can save 26.2%.

Biogas production per month was 90 m³ when converted to LPG 41.4 kg (1 m³ is equivalent to 0.46 kg LPG), the price of LPG measuring 12 kg of 9.85 USD, the price per kg of 0.82 USD. The biogas equality value produced per month compared to LPG was 34.03 USD. Biogas consumption after purification can save 26.07%, or 23.46 m³, with a price of 19.29 USD. The cost of adsorbent material for biogas purification was 2.6 kg zeolite, 2 kg silica gel, and 0.9 kg iron powder 7.37 USD. The cost savings for using biogas after refining amounted to 11.92 USD. The cost of removing impurities was 0.13 USD.

3.8 Biogas testing that has been purified against lighting

Testing of the biogas used as an energy source to ignite using lantern lamps was carried out with the help of air pumps to stabilize the biogas supply to the generator. Table 1 showed the results of testing for biogas consumption to ignite a lantern lamp.

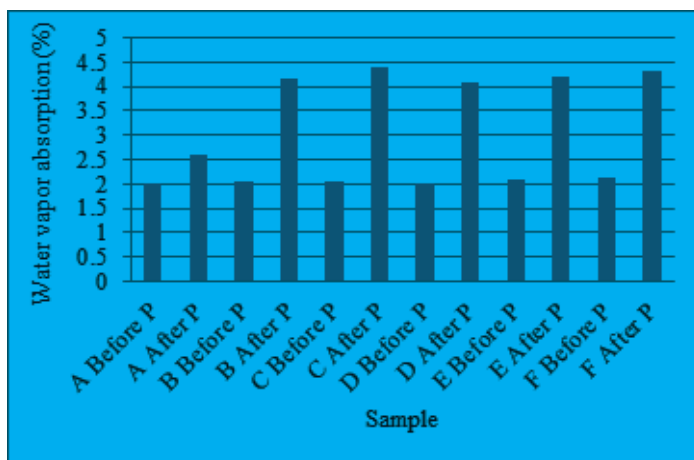


Figure 6. Water vapor absorption before and after biogas purification (P)

Table 1. The result of testing for biogas consumption to ignite lantern lamp

Number of lantern lamp	Biogas consumption before purification (litres/hour)	Biogas consumption after purification (litres/hour)
1 lantern lamp	538.5	394
2 lantern lamp	874	655

Table 2. Test results of biogas consumption to ignite an electric bulb per hour

Electric load bulb lamp (watt)	Biogas consumption	
	before purification (litres/hour)	after purification (litres/hour)
1000	618	452
1100	627	459
1200	639	468
1300	642	470
1400	657	481
1500	667	499
1600	665	494
1700	651	486
1800	645	481

The consumption of biogas to ignite one lantern lamp before purification was 538.5 liters per hour, while after purification was 394 L/hr. The Consumption of biogas to ignite two lantern lamps before purification was 874 L/hr, while after purification was 655 L/hr. Consumption of biogas to ignite two lantern lamps was lower than that of two lantern lamps. The results of the study by Sarifudin (2009) showed that the consumption of biogas to ignite one lantern lamp was 676 L/hr.

The Consumption of biogas to ignite a lantern lamp before purifying biogas was higher than that after purification. Compared to the use of unrefined biogas to ignite one and two lantern lamps, the use of pure biogas can save 26.8% and 25.06% respectively; this was because after purification the content of CH₄ was higher, CO₂ and H₂S content were lower.

Table 2 showed the results of biogas consumption to ignite electric lamp bulbs at various loads. The measurement results determined that there was a difference in the level of use of biogas, which was not

purified and purified in different electrical loads. For load ranges from 1000 to 1500 W, the consumption level of the two types of biogas increased with the increased electrical load. For load ranges from 1600 to 1800 W, the consumption level of both types of biogas decreased with the increased electrical load. Temperature conditions that were not optimal in generator combustion chambers could result in high biogas consumption. At the end of the test, it was likely that the temperature in the generator set combustion chamber was optimal for the combustion process. At the condition where the temperature in the combustion chamber was optimal, biogas consumption would decrease again and showed the same pattern as the initial conditions even though the electricity load was higher; this meant that the level of biogas consumption was not affected by electrical loads since it was below the maximum capable generator (4kW). A previously published paper reported that the use of biogas generators would be more economical when used on large electrical loads (Arfah, 2014)

The consumption of biogas at loads of 1000 - 1800 watts before purification was 618 - 667 L/hr, an average of 645.67 ± 16.42 L/hr, while that after the removal was 452 - 499 L/hr, an average of 476.67 ± 14.77 L/hr. Analysis results of the biogas consumption to ignite the bulb lamps at a load of 1000 - 1800 W showed that the consumption of biogas after purification was lower than that before purification. Compare to unpurified biogas consumption, the use of purified biogas could save 26.3 % of the biogas fuel. The lower consumption of biogas indicated that the purification of biogas increased the methane content from 54.03% to 64.13%, decreased CO₂ from an average of 23.39% to 12.51%, and decrease H₂S from an average of 3641 ppm \pm 543.7 ppm to 0 ppm.

4. Conclusion

The obtained results of this research work could be summarized and concluded as follows;

The maximum percentage of CO₂ removal was achieved in the biogas purification process using zeolite was 52.18%. The use of zeolite adsorbent per gram can remove CO₂ by 0.072%. The rate of H₂S removal in the biogas purification using iron powder was 100%. The use of iron powder adsorbent per gram can remove H₂S by 0.013%. The effectiveness of adsorption of water vapor content in biogas achieved in the biogas purification adsorption process using silica gel was 50.35 g H₂O/m³ biogas. Each gram of silica gel absorbs 0.017% g H₂O/m³ of water vapor.

The maximum percentage of CH₄ increasing obtained in the biogas purification adsorption process was 25.38%, and the percentage in calorific value increasing was a maximum at 11.12 kcal/m³. The consumption of biogas ignites lantern lamps after purification was 26.8% lower than that before purification, and the use of biogas to ignite the lamps bulb after purification was 26.3% lower than that before purification.

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