THE EFFECTS OF ADSORBENT MASS USING RED BRICK POWDER ON THE RESULTS OF BIOGAS PURIFICATION

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THE EFFECTS OF ADSORBENT MASS USING RED BRICK POWDER ON THE RESULTS OF BIOGAS PURIFICATION

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Biogas is produced by the digestion of organic waste by anaerobic bacteria. However, the application of raw biogas is not effective because it consists of impurities such as carbon dioxide (CO\textsubscript{2}), hydrogen sulfide (H\textsubscript{2}S), water vapor (H\textsubscript{2}O), and other impurity gases. Physical Adsorption is the simplest method of immobilization of biomolecules such as CO\textsubscript{2} which is attached to the surface through the weak bonds like van der Waals forces. One of the physical adsorption means to reduce CO\textsubscript{2} levels in biogas is to use brick powder. Increasing of brick powder adsorbent mass was researched at intervals of 5 and 20 minutes, for the CO\textsubscript{2} concentration data and heating values of biogas. The application of 200 and 400 grams brick powder adsorbents, with the biogas flow rate of 1 and 2 liters/minute, was researched at intervals of 5 and 20 minutes, for the CO\textsubscript{2} concentration data and heating values of biogas. Gas Chromatography (GC) was used to determine the concentration of adsorption gases, especially CO\textsubscript{2} and CH\textsubscript{4}. The results showed that the biggest efficiency reduction in CO\textsubscript{2} concentration is 59.28%.

Key words: biogas purification, adsorbent of red brick powder, 2³ factorial design

INTRODUCTION

Renewable energy production is a big issue in the world. With different technologies, a lot of renewable energy can be obtained, such as: solar energy, wind energy, geothermal energy, and biomass energy [1]. Biomass is the main energy resource for biogas as a renewable energy, being mostly found in agricultural residues, livestock manure, and others [2]. Organic households, restaurants, parks, and wastes from industry [3], [4]. Biogas is produced from anaerobic bacterial digestion of degraded organic waste [5]. The composition of biogas consists of 35-75 % methane, 25-65 % carbon dioxide, 1-5 % hydrogen, and tracer of water vapor, ammonia, and hydrogen sulfide. Poor biogas quality (CH\textsubscript{4} / CO\textsubscript{2} < 1) exacerbates damage to the atmosphere [6]. Purification techniques are used to clean and enhance biogas into biomethane which can replace natural gas [7].

Purification of biogas aims to improve its quality by separating non-methane gases, specifically carbon dioxide, which can reduce the heating value and the combustion efficiency [8]. Decreasing the concentration of carbon dioxide can be done by physical adsorption, which is the simplest method to immobilize of biomolecules such as CO\textsubscript{2} which are bounded to the surface through weak bonds such as van der Waals forces [9]. There are three types of adsorbents, including elements that contain carbon (activated carbon and graphite), polymer elements (porous polymer matrix), and elements that contain oxides (silica gel and zeolites) [10].

XRF testing is carried out to find out the elements that exist in a material such as clay by bombarding the material with high energy X-rays or gamma rays so that the waveforms of the clay-forming elements are detected. [11]. Red brick is one of the ceramic products made from clay whose particles are shaped like a sheet that has a special surface with Al\textsubscript{2}O\textsubscript{3} and SiO\textsubscript{2} content in it [12]. Clay has high adsorption ability, it has very strong compressive power, it wrinkles when dried, and it has fine-grained granules so that it can be categorized as montmorillonite [13]. A mixture of tile powder and zeolite can reduce CO\textsubscript{2} concentrations by 34.56 % [14].

RESEARCH METHODS

Red brick powder, which came from the village of Urek - Urek Gondanglegi, Malang, Indonesian. The red brick adsorbent had fine structure, with particle size between 0.0625-0.4 mm. Each adsorption tube was filled with 200 and 400 g of adsorbent. Tests were conducted at biogas flow rates of 1 and 2 Lmin\textsuperscript{-1} and purification times of 5 and 20 min. Biogas is the gaseous result of digesting the chicken and cow manure in anaerobic conditions.

The composition of the red brick content based on XRF test consisted of 55.2 % SiO\textsubscript{2}, 17.1 % Al\textsubscript{2}O\textsubscript{3}, 13.1 % CaO, 4.2 % MnO, 3.2% Na\textsubscript{2}O, 3.14 % TiO\textsubscript{2}, and 2.4 % BaO.

The following parameters were followed: M – adsorbent mass (g), Q – biogas flow rate (Lmin\textsuperscript{-1}) and t – purification time (min). The equipment used in this research are Gas Chromatography (GC) at Greenhouse Gas Laboratory (Balingtan, Pati, Center Java), rotap rocker as a sieve to adjust grain size, digital balance to measure the mass of adsorbent, tedlar bag to accommodate post-purification gas samples, flowmeter to regulate flow rate, stopwatch to measure purification time, polyurethane hose for biogas distribution lines, distribution tubes for storing pressurized biogas, purifying equipment and biogas compression for transferring biogas from digesters.
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Tools and materials were prepared according to Figure 1. Biogas flows through tubes (no. 6 and 7) which have been filled with adsorbents into the sample bag.

**Figure 1: The scheme biogas purification equipment**

Sampling done at time variations of 5 and 20 minutes, the sample bag was fully loaded (2 liters). Two repetitions were made for each variation. After the data obtained, an analysis was carried out using factorial design method $2^3$ to determine the effect of each treatment and the interaction between treatments on the binding of CO$_2$ to biogas purification.

**RESULTS AND DISCUSSION CO$_2$ CONCENTRATION**

The percentage data of the results of CO$_2$ binding using red brick powder adsorbents obtained from Gas Chromatography (GC) are shown in Table 1.

Based on the data in Table 1 obtained a combination of factorial design data $2^3$ as shown in Figure 2.

The use of factorial design $2^3$ was carried out to determine the effect response of each adsorbent mass factor (M), flow rate (Q) and purification time (t) to the percentage CO$_2$ produced by biogas. High level (+) show high treatment value and low level (-1) shows low treatment value on each factor [15][16]. The matrix from in Figure 3 was obtained:

Where:
- y: The percentage of CO$_2$ after biogas purification (%)
- IM: The Mass Factor of Brick Powder Adsorbent (g)
- IQ: The Biogas Flowrate Factor (L min$^{-1}$)
- It: The Purification Time Factor (min)

With the Gauss Siedel iteration method, the completion of matrix on Figure 3 is obtained:

$$y = 14.976 - 4.0198M + 1.039Q - 0.233t - 0.784.MQ - 0.063.MT - 0.0335.Qt + 0.119.MQt$$

Where:
- y: The percentage of CO$_2$ after biogas purification (%)
- IM: The Mass Factor of Brick Powder Adsorbent (g)
- IQ: The Biogas Flowrate Factor (L min$^{-1}$)
- It: The Purification Time Factor (min)

**Table 1: $2^3$ factorial design scheme and % CO$_2$ after biogas purification**

| Coding | M (g) | Q (L min$^{-1}$) | t (min) | (% CO$_2$) | (% CO$_2$) | y (% CO$_2$) |
|--------|-------|------------------|---------|------------|------------|-------------|
| - - -  | 200   | 1                | 5       | 17.25      | 17.13      | 17.189 (1)  |
| + - -  | 400   | 1                | 5       | 10.32      | 11.84      | 11.082 a    |
| - + -  | 200   | 2                | 5       | 20.44      | 21.84      | 21.141 c    |
| + + -  | 400   | 2                | 5       | 12.67      | 10.18      | 11.422 ac   |
| - + +  | 200   | 1                | 20      | 17.18      | 17.13      | 17.154 b    |
| + - +  | 400   | 1                | 20      | 10.33      | 10.31      | 10.319 ab   |
| - + +  | 200   | 2                | 20      | 20.84      | 20.16      | 20.497 bc   |
| + + +  | 400   | 2                | 20      | 10.28      | 11.73      | 11 abc      |
| - - +  | 200   | 1                | 20      | 17.18      | 17.13      | 17.154 (1)  |
| + - +  | 400   | 1                | 20      | 10.33      | 10.31      | 10.319 ab   |
| - + +  | 200   | 2                | 20      | 20.84      | 20.16      | 20.497 bc   |
| + + +  | 400   | 2                | 20      | 10.28      | 11.73      | 11 abc      |

Without treatment 25.34

**Figure 2: Combination of CO$_2$ data on factorial design $2^3$**

**Figure 3: Factorial Design $2^3$ CO$_2$ Data Matrix**
IMQ: The Interaction between Adsorbent Mass and Biogas Flowrate
IMt: The Interaction between Adsorbent Mass and Purification time
IQt: The Interaction between Biogas Flowrate and Purification time
IMQt: The Interaction between Adsorbent Mass, Biogas Flowrate, and Purification Time

Equation (1) explains that the greatest effect (≥ ± 0.5) occurs on the mass factor of adsorbent red brick powder, biogas flow rate, and mass interaction - flow rate on CO₂ levels. A negative value on the M coefficient means that there is an effect of increasing mass on CO₂ levels, CO₂ concentration on 200 grams of the adsorbent mass is higher than 400 grams of the adsorbent mass, which means that the use of the adsorbent mass is more effective with higher levels. Likewise, mass - flow rate interactions have an effect on decreasing CO₂ levels after purification even in small amounts. However, a positive value on the flow rate coefficient indicates that increasing the flow rate does not increase the binding of CO₂ so that the flow rate of 1 L/min⁻¹ is better for binding carbon dioxide compared to 2 L/min⁻¹. This will be proven in the analysis of each treatment variable.

Analysis of the effect of the adsorbent mass on CO₂ concentration in biogas with variable flow rates and time constant be seen in Figure 4. Figure 4 shows that a mass of 200 grams has a higher CO₂ concentration than a mass of 400 grams. The increase in the mass of the adsorbent will reduce the value of CO₂ concentrations as a result of biogas purification. Increasing the mass of the adsorbent means that the volume also increases which will increase the area of the adsorbent. This will result in the increase of the contact area between the CO₂ gas and the adsorbent, so the CO₂ content will continue to decrease along with the increase in the mass of the adsorbent. These results are in line with previous studies [17], [18], [19], [20].

Analysis of the biogas flowrates effect on CO₂ concentrations in biogas with mass and time variables can be seen in Figure 5.

![Figure 4: Adsorbent mass effect on CO₂ concentrations](image)

![Figure 5: The diagram of biogas flowrates effect on CO₂ concentrations](image)
Figure 5 shows that the flow rate of 2 L min\(^{-1}\) has a higher \(\text{CO}_2\) concentration than the flow rate of 1 L min\(^{-1}\). These results are in accordance with previous studies [21], [22] that high flow rates can cause a lot of biogas to be wasted because it was not adsorbed beforehand. Significant differences in \(\text{CO}_2\) concentrations between the flow rates 1 and 2 L min\(^{-1}\) occur at a mass of 200 grams with a delta of around 3%. This happens because the adsorbent is in small amount but the flow rate is too fast so that a lot of biogas is wasted without being bounded into the pores of the adsorbent.

The calculation of the efficiency of \(\text{CO}_2\) binding is done to find out the best treatment, using the formula [23]:

\[
\eta = \left(1 - \frac{\text{CO}_2^{\text{out}}}{\text{CO}_2^{\text{in}}} \right) \times 100\%
\]  

(2)

where:

\(\eta\): The Efficiency of \(\text{CO}_2\) Adsorption (%)

\(\text{CO}_2^{\text{out}}\): The \(\text{CO}_2\) Concentration After Purification (%)

\(\text{CO}_2^{\text{in}}\): The \(\text{CO}_2\) Concentration Before Purification (%)

The results of calculating the effectivity of \(\text{CO}_2\) adsorption are shown in Figure 6.

Figure 6, the highest effectivity occurred in the M400Q1L20m treatment at 59.28%. The lower the concentration of \(\text{CO}_2\) after purification, the value of the effectivity of \(\text{CO}_2\) increases [14], [24]. It happens because the interaction of the variables makes the contact area larger between the red brick powder adsorbent and \(\text{CO}_2\) for the 400 gram mass variable, the binding of \(\text{CO}_2\) with the red brick powder adsorbent is more evenly distributed and minimal biogas is wasted for variable flow rates of 1 L min\(^{-1}\), and the binding of \(\text{CO}_2\) is more maximal and evenly distributed before the saturated red brick adsorbent allows no biogas to be wasted for a 20 minute purification time variable. The interaction of these three variables makes the effectivity of the \(\text{CO}_2\) adsorption with adsorbent red brick powder to be high.

\(\text{CH}_4\) CONCENTRATION

Percentage data of post-purification \(\text{CH}_4\) results using red brick powder adsorbents obtained from Gas Chromatography as shown in Table 2.

Data from Table 2 is converted to a combination of data models as shown in Figure 7.

Table 2: 2\(^3\) factorial design scheme and % \(\text{CH}_4\) after biogas purification

| Coding | M (g) | Q (L min\(^{-1}\)) | t (min) | \(\%\) \(\text{CH}_4\) | \(\%\) \(\text{CH}_4\) | \(\%\) x |
|--------|-------|----------------|--------|----------------|----------------|-------|
| - - -  | 200   | 1             | 5      | 47.53          | 48.35          | 47.936 (1) |
| + - -  | 400   | 1             | 5      | 58.68          | 67.13          | 62.904 a  |
| - + -  | 200   | 2             | 5      | 66.32          | 56.98          | 61.648 c  |
| + + -  | 400   | 2             | 5      | 64.44          | 65.00          | 68.402 ac |
| - + +  | 200   | 1             | 20     | 69.09          | 45.10          | 57.096 b  |
| + + +  | 400   | 1             | 20     | 67.42          | 66.44          | 66.929 ab |
| - + +  | 200   | 2             | 20     | 47.33          | 59.10          | 53.213 bc |
| + + +  | 400   | 2             | 20     | 69.04          | 67.76          | 65.717 abc |

Without treatment 36.83

![Figure 7: Combination of \(\text{CH}_4\) Data on Factorial Design 2\(^3\)](image)
The use of factorial design $2^4$ was carried out to determine the effect response of each adsorbent Mass (M), Flowrate (Q) and Purification Time (t) to the % CH$_4$ of biogas purification. High level (+1) shows high treatment value and low level (-1) shows low treatment value on each factor [15]. The matrix form is obtained in Figure 8; Method of the Gauss Siedel iteration was used the completion of matrix on Figure 8 is obtained:

$$
\begin{array}{cccc}
1 & -1 & -1 & 1 \\
1 & 1 & -1 & -1 \\
1 & 1 & 1 & 1 \\
1 & 1 & 1 & 1
\end{array}
$$

\[ F_0 \]
\[ F_M \]
\[ F_t \]
\[ F_{MQ} \]
\[ F_{MQt} \]

\[ \begin{array}{c}
47.936 \\
62.904 \\
61.648 \\
68.402 \\
57.096 \\
66.929 \\
53.213 \\
65.717
\end{array} \]

Figure 8: Factorial Design $2^4$ CH$_4$ Data Matrix

where:
- $x$: The percentage of CH$_4$ after biogas purification (%)
- $F_M$: The Mass Factor of Brick Powder Adsorbent (g)
- $F_t$: The Biogas Flowrate Factor (L min$^{-1}$)
- $F_{MQ}$: The Interaction between Adsorbent Mass and Biogas Flowrate
- $F_{MQt}$: The Interaction between Adsorbent Mass, Biogas Flowrate, and Purification time
- $F_{MQt}$: The Interaction between Adsorbent Mass, Biogas Flowrate, and Purification time

Equation (3) explains that the greatest effect (≥0.5) occurs in the mass factor of the adsorbent red brick powder, biogas Flowrate, Mass interaction - Flowrate, Flowrate - Purification Time Interaction, and Mass - Flow rate - Purification Time Influence CH$_4$ levels. A positive value on the M coefficient means that there is an effect of increasing Mass on CH$_4$ levels [17], [19], [20]. Increasing the mass of the adsorbent will increase the percentage of CH$_4$ because a lot of CO$_2$ is bound to the adsorbent, thereby increasing the ratio between CH$_4$ and CO$_2$.

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