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Experimental investigation on the simultaneous upgrading of biogas using iron sponge and zeolite 13x fixed bed columns

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Abstract

Biogas technology is continuously being studied to enhance its quality and expand its power generation application. To increase its heating value and eliminate its corrosive effect on machine components, a biogas upgrading system was designed and developed. This study evaluates the developed upgrading system by its capability to increase the methane content through hydrogen sulfide and carbon dioxide reduction using the iron sponge and Zeolite 13x, respectively as adsorbent columns. The results show that by varying the volume flow rate of the biogas into the system could lengthen the contact time between the biogas and adsorbent and eventually increase the CH_4 content from the biogas. Furthermore, the highest methane content achieved at 63.1 % from a raw biogas concentration of 32% CH_4. Meanwhile, with the breakthrough adsorption capacity of zeolite 13x was 224 mg CO_2/g. This had reduced the CO_2 by 41.76% from its raw biogas composition. The maximum superior and inferior calorific values produced were 22.53 MJ/m³ and 20.29 MJ/m³ respectively. The simplified design of a simultaneous upgrading system intends to increase the methane content of the biogas and further extend its application as an alternative fuel for power generation applications.

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Introduction

Biogas, as an alternative source of energy, can be obtained relatively easily from plants and animals including nutrients, excrement, and bio-waste from households and industry. It has been recognized as a clean and renewable energy source that can replace fossil fuels, thus mitigating the environmental problems caused by the latter. The transformation of biomass to biogas involves a process called anaerobic digestion. It encourages microorganisms to digest the organic material to produce a gas rich in methane and carbon dioxide. The four fundamental steps of anaerobic digestion are hydrolysis, acidogenesis, acetogenesis, and methanogenesis (Biarnes, 2018). The approximate biogas composition after anaerobic digestion varies from every substrate, but the values range from 45-70% CH4, 30-45% CO2 and small amounts of other components such as H_2O and H_2S and a relatively small amount of hydrocarbons (Rasi, 2009).

Furthermore, the basic knowledge in biogas production: the biochemical processes, the storage, and purification technologies and the application improvements are being widely extended. The recent configurations and integration that were applied to improve these processes resulted in a substantial increase in biogas production. The removal of unwanted compounds from biogas streams has been of considerable importance in the past and is even more so today. Feasible biogas purification technologies already exist to upgrade the efficiency or the calorific value of the biogas.

The overall calorific value of the gas mixture is highly dependent on its methane content. Pure methane has an upper calorific value of 39.8 MJ/m^3, which corresponds to 11.06 kWh/m^3 (Jørgensen, 2009). However, the CO_2 and other impurities present in the biogas are noncombustible. These unwanted compounds cause a detrimental effect on the heating value of the biogas when burned as fuel. Traditionally, biogas is directly burned in a stove for cooking. But with the use of purification, biogas is being upgraded to natural gas standards such as bio-methane and be used as fuel for power generation.

Zeolites, commonly referred to as molecular sieves are highly porous and especially effective at removing polar compounds such as water and H_2S from non-polar gas streams like methane. Zeolite 13x with pore size approximately 8 Å has the largest adsorption capacity for undesirable components in comparison with the other adsorbents. As one of the simplest methods, iron oxides remove hydrogen sulfide by forming insoluble iron sulfides. Steel wools have high porosities- 75-95% of its volume are cellular structures made with a large volume fraction of pores. This allows the passage of gas through its surface area that makes it ideal adsorbent for physical adsorption.

However, biogas in its raw state does not only have low heating value but also poses significant problems to machine components when used as a fuel. That leads to the study emphasizes on the need to treat biogas under CO_2 and H_2S removal systems using Zeolite 13x and Steel wool to provide data for the effectiveness of the recent configurations and integration of physical adsorption in the biogas upgrading technology. By developing a simplified design of a simultaneous upgrading system, the system is intended to increase the methane content of the biogas and further extend its application.

Methodology

This is an active pursuit to increase the performance of the biogas in terms of its quality and range of application by experimentally providing data for the effectiveness of physical adsorption in the biogas upgrading technology. By removing the impurities in the gas mixture, the methane ratio, which is the basis for its fuel heating value will be increased. It is then imperative that its potential as a primary source of energy for power generation will also improve.

This study will determine the performance of a two-stage simultaneous upgrading system. Two fixed bed reactors utilizing the principle of physical adsorption using Zeolite 13X beads and Iron oxide from the iron sponge. The experimental testing will be conducted through lab-based manual data gathering. The design parameters will be recorded before and after the tests for a comparative assessment.

Slurry preparation

35 kg of fresh cow manure is weighed and mixed with 52.5 L of water (1:1.5). The mixture is stirred thoroughly and the solid materials like pebbles were removed until homogeneity is achieved. The mixture is poured inside the 100 L anaerobic digester and the valve connecting it to the gas storage is opened. The digester is regularly stirred through an agitator daily. The digester is allowed to produce gas within 30 days of retention time and stored in the floating drums for testing.

Steel wool preparation

200 ml of Hydrochloric acid is poured inside a container. Using protective gloves, 100 g of iron sponge (steel wool) is soaked in the solution for about 1 minute. The steel wool is drained and the remaining solution is removed by wrapping it in a thick dry cloth. Upon drying, the steel wool is sealed inside the column.

Biogas upgrading system process

Raw biogas is stored in a floating drum to supply the upgrading system. The initial composition, volume flow and temperature of the biogas are taken at the inlet section of the iron sponge column using a digital thermocouple and water manometer. A gas flow meter is installed to control the flow of the biogas to the system.

The raw biogas is then gradually fed into the 1st stage iron sponge packed-bed at a 200 ml volume flow rate. The biogas passes through the 2nd stage packed-bed column leaving the CO_2 adsorbed in the surfaces. Upon exiting the Zeolite reactor, another gas analysis is done and the same set of parameters is then recorded. These three sets of data are to be compared and analyzed to evaluate the effectiveness of the system. The temperature inside the fixed-bed columns should be maintained within the range of 25 degree Celsius or a little bit higher (Siefers, 2010). Fig.1 shows the process flow for the upgrading system, while Fig. 2 represents the biogas schematic diagram of the system, as well as the actual set up in Fig. 3, and the developed upgrading systems shown in Fig. 4.

The steel wool column for the H_2S removal system is positioned vertically as prescribe (Siefers, 2010) with flow going downwards so that the gas passes through the most fouled bed first. The Zeolite 13X column, on the other hand, is positioned horizontally to reduce compaction because no mechanical device was used to force the gas through it. Several ball valves were installed in between columns and in every sampling point for convenience in the gas sample extraction. 10 mm diameter nylon pneumatic hose was used for the connections because it was specified to work on pressure ranging from 0.5 MPa to 5 MPa and -40^{o}C to 93^{o}C temperature range.

Results and discussion

Effects of adsorption capacity on the simultaneous upgrading using Iron sponge and zeolite 13X adsorbent

Figure 5 shows the data for the test with 200 ml/min flow, the gas was able to reach the gas storage within 8-11 min duration and a significant increase in the methane content. Unlike the first two trials, the biogas was already purified in the first twenty minutes of the experiment.

The highest methane content recorded was 63.1% which doubles the percentage of the raw biogas fed into the system which is only 32 % CH_4.

The mass of zeolite 13x and steel wools were 500 g and 100 g respectively in each test. The results show that, though hypothesized that compaction will improve adsorption capacity, it hindered the flow of gas through the columns since no mechanical devices were used to force the gas through it. When the Zeolite 13x column was installed vertically, it increased the compaction and limited the flow of gas. Although it increases the contact time, it produces minimal purified gas output.

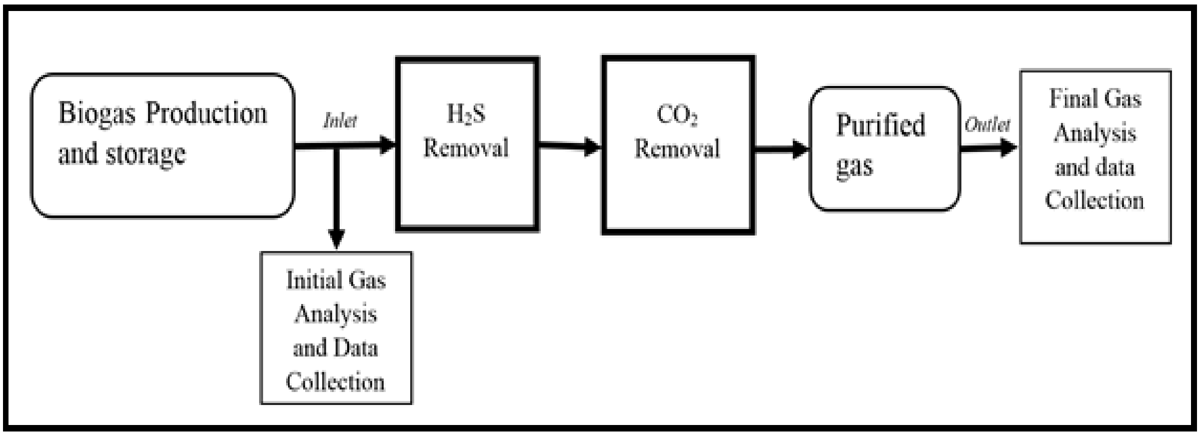

Fig. 1. Biogas upgrading system process flow.

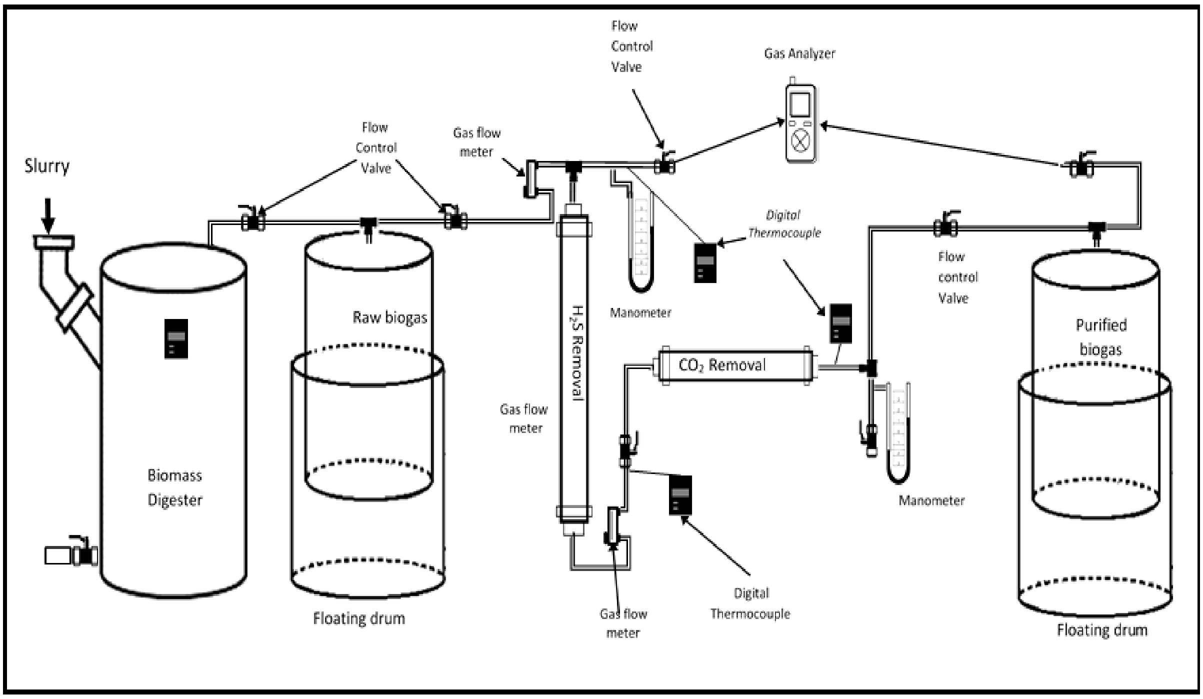

Fig. 2. Schematic diagram of the biogas upgrading system.

The pressure is directly proportional to the volume flow of the gas. A minimal increase in pressure was observed and remained constant in each trial. It was observed during the experimental trial that 300 ml/min with higher pressure compared to 100 ml/min resulted in a slightly higher methane content.

The temperature record during the experiment was almost identical to the ambient temperature. Although the temperature from the digester was higher, it was reduced to ambient temperature when the gas was stored inside the floating drum. There will be no significant effect on the methane content if the temperature is varied (Siefers, 2010). Hence, the temperature is maintained to 25 ^{o}C and above as prescribed.

Breakthrough analysis of zeolite 13X

Longer contact time was hypothesized to have a significant effect on the adsorption capacity of the adsorbents until the time of observed breakthrough. The bed contact time was varied by adjusting flow to the adsorbent columns using a gas flow meter. The flow was initially set to 300 ml/min (102.1053 kPa) but results after the upgrading showed little increase in the methane content which probably due to shortened contact duration. As shown in Fig. 6, the methane content was very low (16.5%-23%) in the first 20 minutes and reached 53.2% CH_4 after an hour. It is because the higher biogas flow rate had negatively impacted contacting efficiency and mass transfer of the gas phase as explained by Fernández (2014) in his journal about rheologically complex fluids.

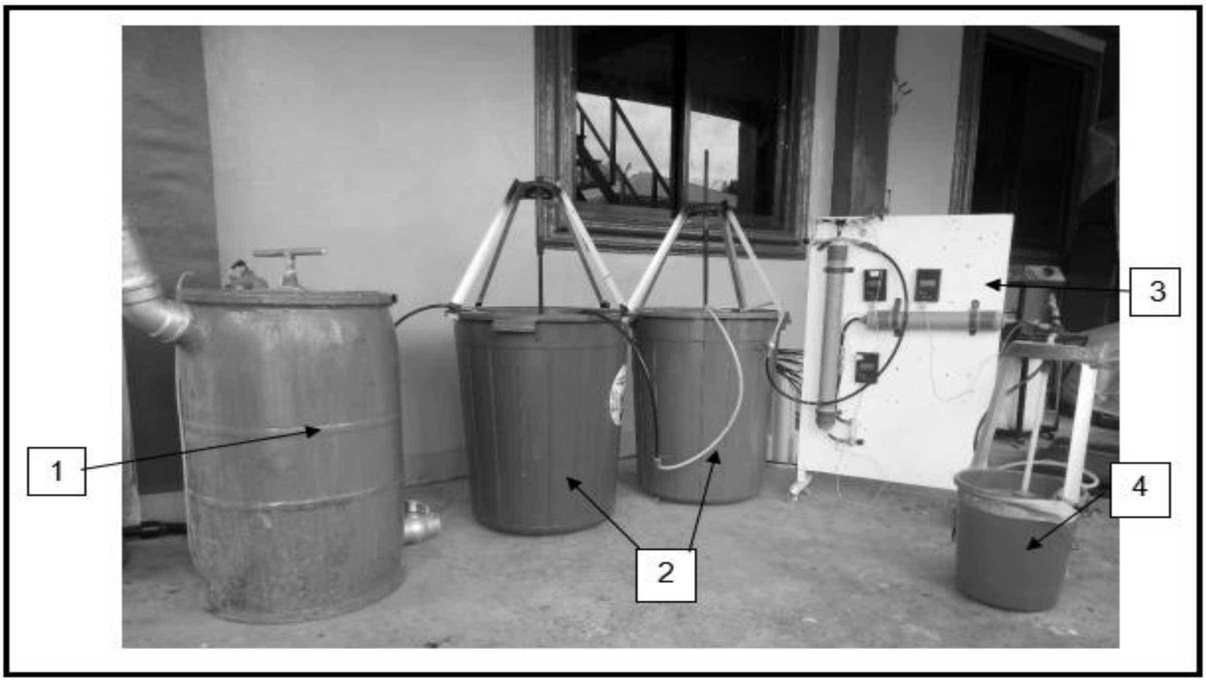

Fig. 3. Upgrading system with digester and storage.

Legend: (1) Digester, (2) Raw Biogas Storage, (3) Upgrading System, (4) Purified Biogas Storage.

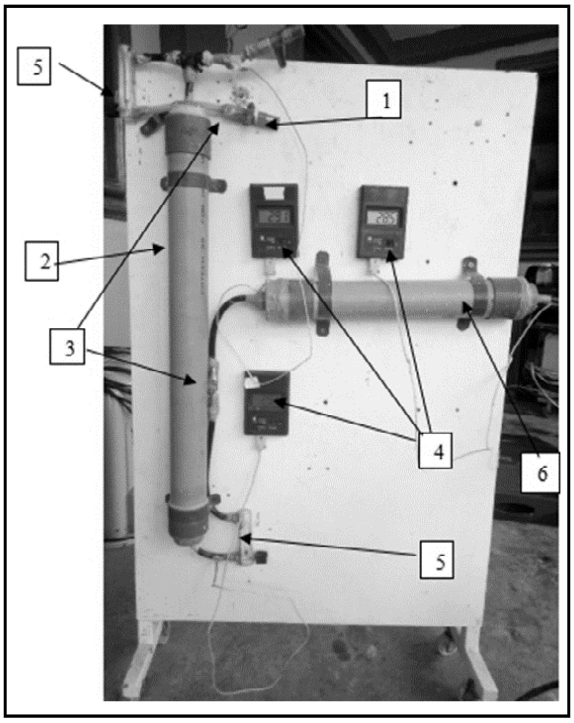

Fig. 4. Developed Biogas Upgrading system.

Legend: (1) Inlet, (2) Steel Wool Column, (3) Flow control valve, (4) Digital Thermocouple, (5) Gas Flowmeter, (6) Zeolite 13x Column

When the gas flow rate was reduced to 100 ml/min, the gas could not easily pass through the zeolite 13x column due to its compaction.

It was observed that lower volume transfer resulted in lower pressure in the system (101.6176 KPa) and according to Le-Chatelier's Principle in case of physisorption of gases over solids, the extent of adsorption increases with increase in pressure as the volume of the gases decreases during adsorption (Principles of Adsorption by Langmuir). Which also led to a very small amount stored in the purified gas storage.

Zeolite adsorption capacity in the breakthrough point (W_{break}), as shown in Fig. 6, was calculated using the breakthrough curve according to the following expression:

$$W_{break} = \frac{U_0 \, C_0 \, t_{break}}{L_t \, \rho_l} \quad \text{(Eq. 1)}$$

$$t_{break} = \int_0^t \left(1 - \frac{C}{C_0}\right) dt \quad \text{(Eq. 2)}$$

t_{break}= 50.11 min
W_{break}= 224 mg CO_2/ g zeolite 13x

The calculated result for the adsorption capacity of zeolite 13x in breakthrough point which is 224 mg CO_2/g zeolite 13x is slightly smaller compared tothe value presented by Alonso-Vicario (2010) in her study which is 298.5mg CO_2/g Zeolite 13X. This result, however, reduced the CO_2 by 41.76 % from its original CO_2 content.

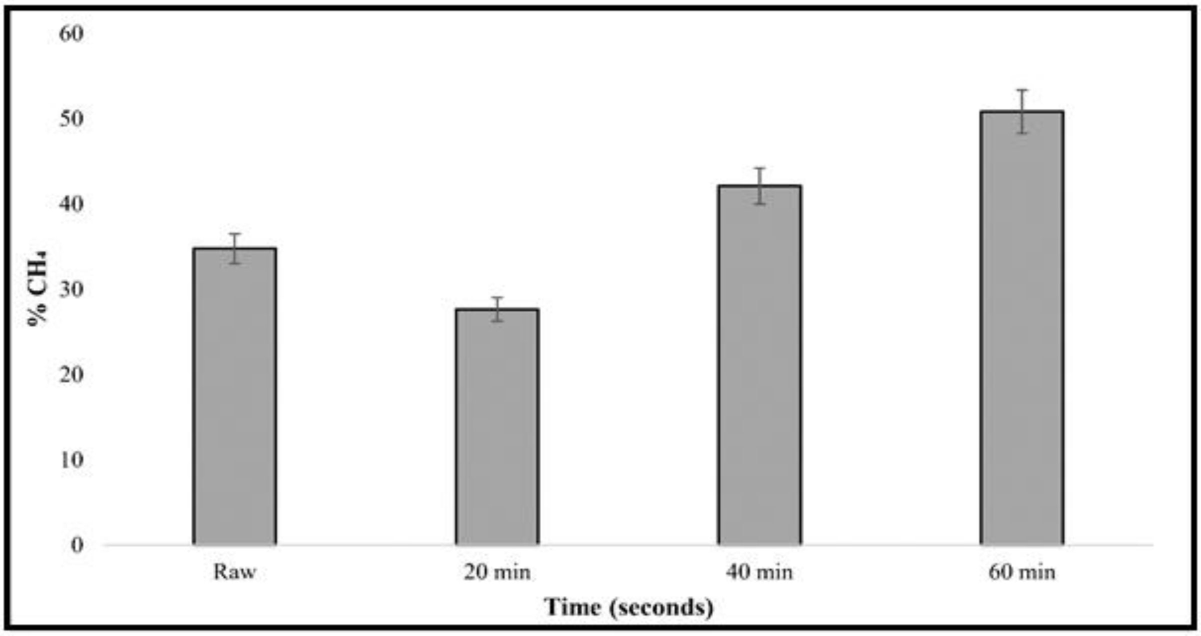

Fig. 5. Biogas methane content.

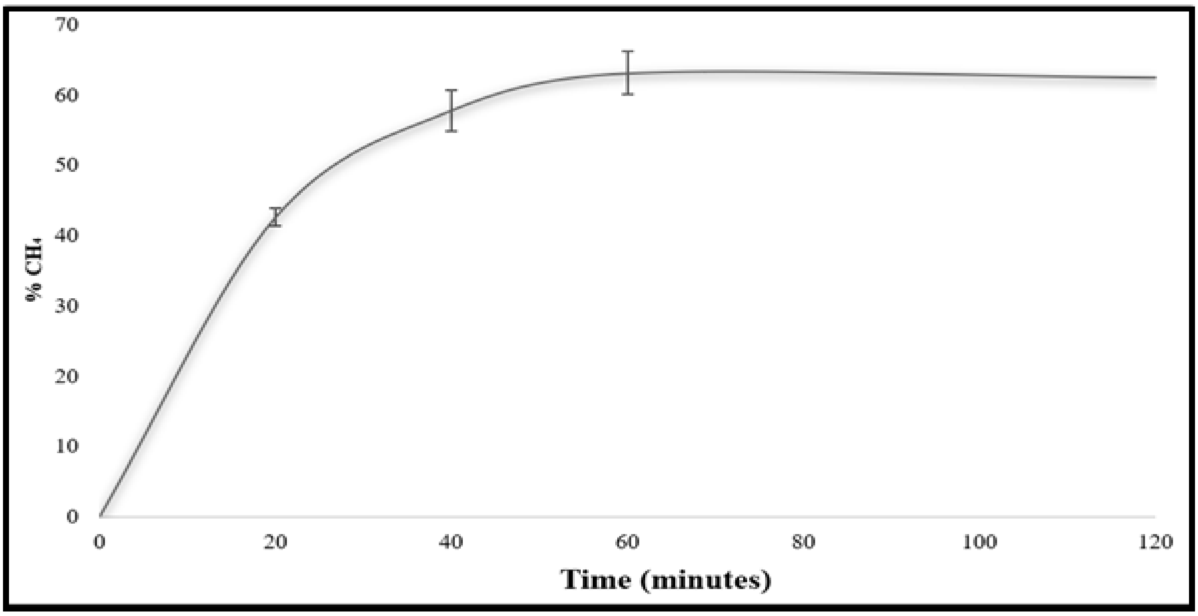

Fig. 6. Breakthrough curve of zeolite 13X.

Calorific (Energy) value of upgraded biogas

The heating values in each trial were calculated using ISO 6976 (1995) Natural gas calculation of calorific values, density, relative density, and Wobbe index from composition and are presented in Fig. 7. The maximum superior and inferior calorific values produced were 22.53 MJ/m³ and 20.29 MJ/m³ respectively. The higher the methane content in the biogas, the higher the heating value will be. The said values are similar as presented by Ludington, 2013.

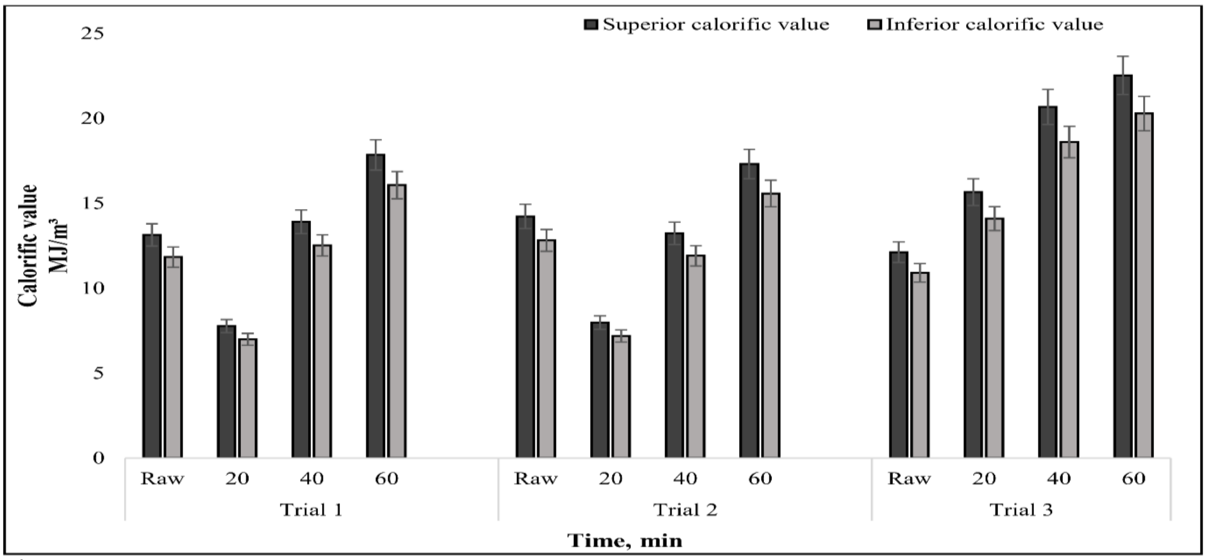

Fig. 7. Superior and inferior calorific value of the biogas.

Conclusion

The need to upgrade biogas under CO_2 and H_2S removal systems aims to increase the methane content of the raw biogas and further extend its application. The designed and developed biogas upgrading system is capable to determine the adsorption capacity of steel wool and Zeolite 13X. This resulted in improved performance of the upgrading system by adjusting the gas flow rate, as well as to increasing methane content. Utilizing varied agricultural biomass wastes as slurry material are recommended to produce a more comprehensive result. Also, recommended to using precise gas measuring instrument to specifically evaluate the changes in the biogas composition during the upgrading process.

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