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ORIGINAL RESEARCH ARTICLE

Performance efficiency of locally available low-cost adsorbents in purification of biogas for high grade applications

Victor O. Odero[1], Francis X. Ochieng'[1], Paul M. Njogu[1]

[1]*Institute of Energy and Environmental Technology (IEET), Jomo Kenyatta University of Agriculture and Technology (JKUAT), Nairobi, Kenya*

Corresponding author email: vodero5@gmail.com

ABSTRACT

Despite having several benefits, the uptake of biogas as a green fuel in rural areas has remained low. One of the technical challenges responsible for slow biogas adoption rate in rural households is biogas contamination. Biogas requires treatment before use, this includes; purification and upgrading. Purification involves the removal of impurities whereas upgrading aims to convert the biogas to a higher fuel standard by increasing its low calorific value. The aimof this study was to develop and test the performance of a cost-effective and affordable system for purification of biogas using low-cost absorbents, thus upgrading biogas to a sustainable clean cooking fuel. The study employed an experimental research design. A pilot domestic scale biogas cleaning system was set up at the Jomo Kenyatta University of Agriculture and Technology (JKUAT) IEET workshop. The study sought to determine the potential and efficiency of locally available and low-cost adsorbents in purification of raw biogas. The raw biogas was passed through the cleaning units, each packed with individual adsorbent. Analysis ofthe biogas for methane (CH_4) composition was done before and after each cleaning unit using digital biogas analyzer model GASTiger 2000, supplied by Yantai Stark Instrument Company limited. The locally available low-cost adsorbents used in the study were; red soil, charcoal, steel wool, clay soil, iron shavings and wood ash. This study also tested the effect of the various physical interfaces of the adsorbents, where a cleaning unit packed with two or more of the adsorbents was used. The study shows that adsorption by clay soil and charcoal gave highest methane concentrations at 67.48±2.33% and 67.20±1.08% respectively from 58.47±2.12% in raw biogas. Cleaning by the other adsorbents; red soil, wood ash, steel-wool and iron shavings improved methane concentrations to 61.76±0.71%, 60.75±2.47%, 58.97±1.92% and 58.56±1.96% respectively. Combination of the various adsorbents into one cleaning unit further improved methane levels to 69.83±1.57%. These results, despite exhibiting relatively low adsorption of the impurities in raw biogas, positively confirmed that indeed the low-cost adsorbent materials have a potential in biogas purification. Modification of these low-cost adsorbent materials, such as use of activated charcoal, would be one of the ways to improve their performance. However, this study specifically sought to explore the potential of these low-cost adsorbents in purification of raw biogas, in their natural state.

This study recommends further exploration of other relatively less technical methods such as water scrubbing that could be coupled with the use of natural low-cost adsorbents to improve the purification

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of raw biogas to near, if not equivalent to, natural gas (that is in the regions of 97-100% CH_4 concentration). Thus, for a possible adoption as an alternative means of cleaning and upgrading raw biogas to a clean cooking fuel.

Key words: low-cost adsorbents, cleaning unit, physical interfaces, biogas purification

1.0 Introduction

Conventional cooking methods such as use of solid fuels, is one of the major sources of environmental pollution. Furthermore, domestic use of these inefficient cooking fuels is a major source of household air pollution (HAP) which has been highlighted by World Health Organization (WHO) as one of the respiratory disease causes (Chafe *et al*., 2014, Lacey *et al*., 2017). A step up and adoption of improved cooking technology is key to mitigating these burdens. If done properly – that is, by ensuring use of clean cooking technologies that offset use of traditional, polluting stoves and fuels – and sustainably, the introduction of clean cooking technology can drive progress towards at least four of the 2030 Sustainable Development Agenda's Sustainable Development Goals (SDG 3,7,13 and 15) (Rosenthal, *et al*., 2018).

When clean cooking programs focus on fundamentally clean fuels, such as LPG, electricity, biogas, or ethanol, available evidence from previous studies suggests that health improvement outcomes are most likely (Lekavičius et al., 2019). Of the available options as clean cooking fuels, biogas stands out as one that is relatively more affordable and less sophisticated to set up its production and utilization, both in the urban and rural settings. It is not noxious; it is colorlessand odorless and is an ideal fuel that can be used for a variety of applications, such as cooking, lighting, and power (Lekavičius et al., 2019). Apart from being a high-quality cooking and lighting fuel, an integrated biogas and sanitation program offers several direct and indirect benefits, including time savings associated with fuel wood collection and cleaning, as well as environmental benefits through the reduction of local deforestation.

There is immense potential and opportunities for biogas, however, lack of adequate; infrastructures, capital, sustainability, awareness, education and appropriate policy, are just some of the challenges that have hindered its successful adoption and usage both at small-scale (household or domestic) and large-scale levels for energy generation, electricity generation, and transportation (Patinvoh &Taherzadeh 2019). In addition to these, biogas purification has also stood out as one of the main challenges facing its usage.

Biogas is generated from organic matter by anaerobic digestion where microorganisms break down the raw materials to produce biogas as a metabolic side product (Kumar et al., 2013). From production, biogas consists majorly of methane and carbon-dioxide. In addition, it contains trace components that include hydrogen sulfide (H2S), ammonia (NH3), nitrogen (N2), hydrogen (H2), oxygen (O2). (Mulu, M'Arimi & Ramkat, 2021). These trace components and carbon-dioxide reduce the energy content of biogas, thus limiting its application. The biogas methane content is in the range of 40-65% with

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calorific value lower than 25MJ/m^3. At standard temperature and pressure (STP) conditions, the calorific value of methane is 36MJ/m^3 (Angelidaki et al., 2018).

Several modern technologies of biogas purification have been explored and reported in various studies. These technologies include; pressure swing adsorption (Fouladi, 2020), cryogenic separation (Yousef et al., 2016); Pellegrini et al., 2018), membrane separation (Makaruk et al., 2010), water scrubbing (Läntelä et al., 2012); Vilardi et al., 2020), physical scrubbing, chemical absorption (Abdeen et al., 2018), hydrate formation (Di Profio et al., 2017) and biological conversion process (Kougias et al., 2018). Other methods include; membrane-based gas permeability, electrical swing adsorption, photosynthetic biogas uptake technique and scrubbing with chemically active liquids.

These commercial upgrading technologies are generally effective in capturing carbon dioxide and trace components in biogas. However, according to Karne et al., these technologies are relatively costly; both the initial investment and operation costs, thus not easily accessible to the many potential domestic biogas users. The process of biogas upgrade using modern technologies has also been reviewed (Ryckebosch, *et al*., 2011).

To promote sustainable development and environmentally friendly technologies, biological biogas purification technologies can be applied (Das, Ravishankar & Lens, 2022). Documented literature, from previous studies, indicate that low-cost natural adsorbents have a potential in purification of biogas through surface adsorption and wet carbonation processes (Mulu, M'Arimi and Ramkat, 2021). However, there is still a gap and a need to investigate methane-loss during the upgrading process using low-cost adsorbents. Comparative investigations of the process cost-effectiveness of using natural materials to upgrade biogas should also be carried out to determine their suitability against the commercial processes (Mulu, M'Arimi and Ramkat, 2021).

The objective of this study was to develop and analyze the performance of an affordable systemfor cleaning of biogas; to determine the potential and efficiency of low-cost adsorbents in purification of biogas.

2.0 Materials and methods

2.1 Materials

The low-cost adsorbent materials used in the study were as discussed below;

2.2 Clay soil

Clay is a fine grain natural rock composed majorly of hydrous aluminum phyllosilicates, with alayered crystal structure. There are two general categories of clay; anionic clay and cationic clay. Though a rare one in occurrence, anionic clay can be synthesized in the laboratories, and it is composed of layered double hydroxides. Anionic clay includes compounds like pyroaurite andhydrotalcite. On the other hand, cationic clay is largely cum readily available in nature and is composed of aluminosilicate layers.

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These include compounds like sepiolite, bentonite and montmorillonite. (Mulu et al., (2021)).

According to Mulu et al., (2021) due to its chemical composition, clay soil is able to remove carbon-dioxide from raw biogas by either physical or chemical adsorption. Clay minerals, suchas kaolinite or montmorillonite, have surfaces with charges that can attract and hold certain molecules, including CO_2, through Van der Waals forces and other interactions.
The process can be represented as below:

Clay (adsorbent) + CO_2 (adsorbate) → Clay-CO_2 (adsorption complex)

Chemically, clay is able to eliminate CO_2 from raw biogas through capture due to its chemical components which include; SiO_2, CaO, Fe_2O_3, MgO, TiO_2, Na_2O and K_2O. Example of CO_2 capture reaction by MgO, which is one of the chemical components of clay, is as shown below;

$MgO+CO_2 \rightarrow MgCO_3$……………………………………………………………………………………………Equation 1

The resulting magnesium carbonate is a solid, thus deposits in the adsorbent material, while the purified gas is pumped to collection bag.

2.3 Iron shavings

These are small iron cut offs, majorly found in iron-dealing workshops or iron commodities' fabrication points. The purification of raw biogas using iron shavings typically involves the removal of hydrogen sulfide (H_2S), which is a common impurity in biogas. Iron is known to react with hydrogen sulfide to form iron sulfide (FeS), a solid precipitate that can be easily separated from the gas stream, thereby purifying the biogas. Below is an equation of the reaction;

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$Fe(s) + H_2S(g) \rightarrow FeS(s) + H_2(g)$……………………………………………………………………………………..Equation 2

In this reaction, iron (Fe) reacts with hydrogen sulfide (H_2S) to produce iron sulfide (FeS) andhydrogen gas (H_2)

2.4 Steel-wool

Steel-wool, also known as wire wool or iron wool, is a bunch of very fine and flexible steel filaments. It is made from low carbon steel in a process where a heavy steel wire is pulled through a toothed die that removes thin, sharp, wire shavings. It is used both commercially anddomestically as a polishing material for woodworks, jewelries and cooking pans.

The purification of raw biogas using steel wool is another method for removing hydrogen sulfide(H2S), a corrosive and toxic gas present in biogas. Similar to iron shavings, as discussed above, steel wool is reactive with hydrogen sulfide leading to a chemical reaction that forms iron sulfide(FeS). Additionally, steel-wool provides a larger surface area for the reaction

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2.4 Wood-ash

Generally, wood ash is generated from the combustion of wood industrially or domestically for power and home fire respectively. Chemical composition of wood ash includes SiO_2, CaO, Fe_2O_3, K_2O, MgO, Na_2O, SO_3, P_2O_5, TiO_2, MnO and Al_2O_3 (Mulu et al., (2021). It also has high contents of carbonates such as potassium carbonates, magnesium carbonates and calcium carbonates. CO_2 can be captured by wood ash in two ways; physical adsorption (similar to physical adsorption of CO_2 by clay) and carbonation. For carbonation process, free calcium oxide (in presence of water) reacts to form calcium hydroxide which then reacts with carbon-dioxide to form calcium carbonate and water as shown in the two equations below;

$CaO + H_2O \rightarrow Ca(OH)_2$……………………………………………………………Equation 3

$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$……………………………………………………Equation 4

In addition to this, wood ash contains alkaline compounds that can react with acidic gases, forming solid compounds that can be easily separated from the biogas. The major acidic impuritygas in raw biogas is hydrogen sulfide. The reaction involves its neutralization:

$H_2S + Ca(OH)_2 \rightarrow CaS + 2H_2O$………………………………………………………Equation 5

2.6 Red soil

As the name suggests, red soil comprises of thin organic and organic-mineral layers of leached soil on a red layer of alluvium. Their red color varies from reddish brown to reddish yellow due to their high iron content, which is in the form of iron oxides or hydrated oxides. Red soil is generally from weathered crystalline and metamorphic rock.

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According to (Mrosso et al., (2020), the use of red soil for the purification of raw biogas is aimed at removing acidic impurities such as H_2S. It is rich in minerals like iron and aluminum oxides, that can react with acidic gases to form solid compounds that can be separated from the biogas. The chemical reaction with H_2S is as below;

$H_2S + Fe_2O_3 \rightarrow Fe_2S_3 + 3H_2O$……………………………………………………………………………………Equation 6

In this equation, iron (III) oxide (Fe_2O_3) is present in red soil. Hydrogen sulfide reacts with iron (III) oxide to produce iron (III) sulfide (Fe_2S_3) and water (H_2O). The iron sulfide is a solidprecipitate that can be separated from the purified biogas. Periodically, the spent red soil may need to be replaced or regenerated for continued efficiency.

2.7 Charcoal

This is the impure form of graphitic carbon, and is obtained as a residue when carbonaceous material is partially burned, or heated under limited air availability. It is also a high surface area compound, exhibited by its molecular structure as below;

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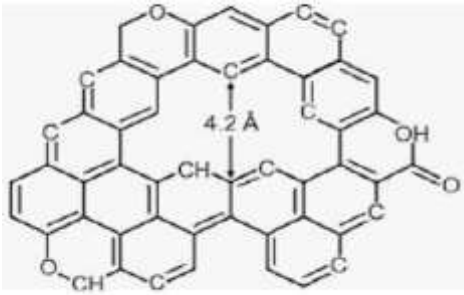

The purification of raw biogas using charcoal targets the removal of impurities such as CO_2, hydrogen sulfide (H_2S) and other trace contaminants. Being a porous material with a large surface area, charcoal is a suitable adsorption material for certain gases.

The main purification process involving charcoal is adsorption rather than chemical reactions. Below are simplifiedrepresentation of the adsorption of H2S and CO2 by charcoal:

H2S (g) + Charcoal (adsorbent) → Charcoal-H_2S (adsorbed)..Equation 7

CO_2(g) + Charcoal (adsorbent) → Charcoal-CO_2 (adsorbed)..Equation 8

In this equation, the hydrogen sulfide gas is adsorbed onto the surface of the charcoal, forming a complex where H_2S is physically bound to the charcoal. This process relies on the porous structure of charcoal, which provides ample surface area for the adsorption to take place.

The effectiveness of charcoal in removing impurities is based on its ability to adsorb gases through Van der Waals forces and other interactions. Once the charcoal becomes saturated withimpurities, it may need to be replaced or regenerated to maintain its purification efficiency.

3.0 Methodology

The research study employed experimental design, both qualitative and quantitative. A biogas cleaning system and process was built, as in the diagram below, and installed at the JKUAT IEET workshop. Attention was paid to the joining points, and proper sealing done, to ensure nogas leakages during the study. This was done and confirmed using soapy water to test for any leakages before commencement of the experiments.

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Figure 2: Fabricated structure

The various natural adsorbent materials i.e. charcoal, clay soil, red soil, steel-wool, iron shavingsand wood ash; were sourced from within and around JKUAT area and prepared for packing into the gas cleaning system. Majorly, the materials' preparation involved size reduction to increase their surface area for adsorption and sizing steel-wool bundles to fit into the system unit pipe.

The adsorbent materials were then packed into the biogas cleaning units, individually, and thegas analyzed using biogas analyzer before and after passing through each of the materials.
Additionally, the various adsorbent materials were packed into one cleaning unit and thepercentage methane measurements done through the same.

The figures below show the design and array of the materials' packing

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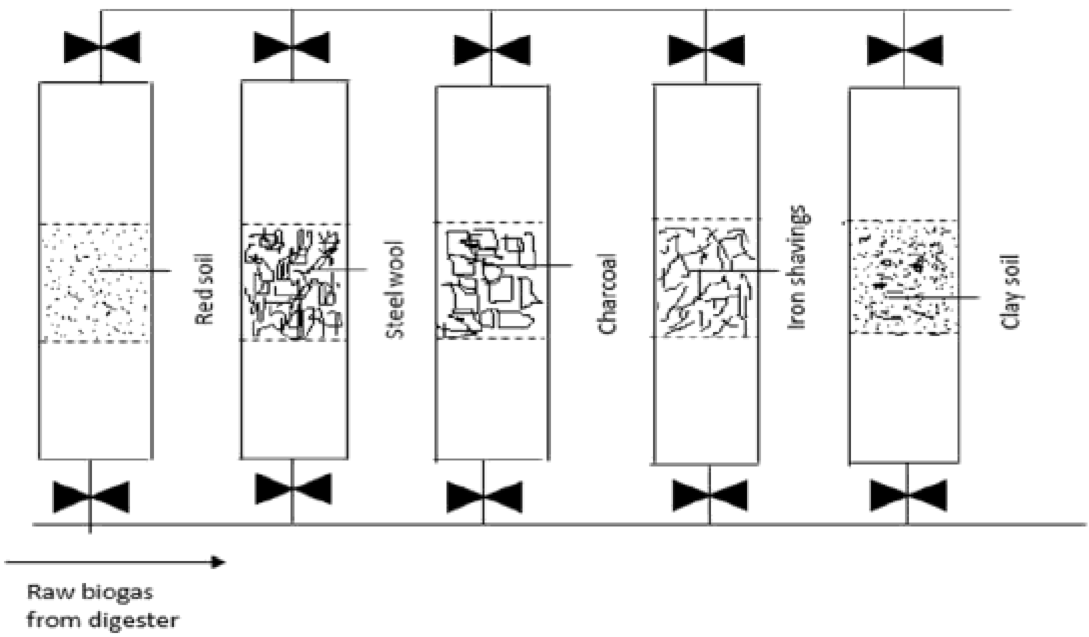

Figure 3: Array of individual adsorbent materials

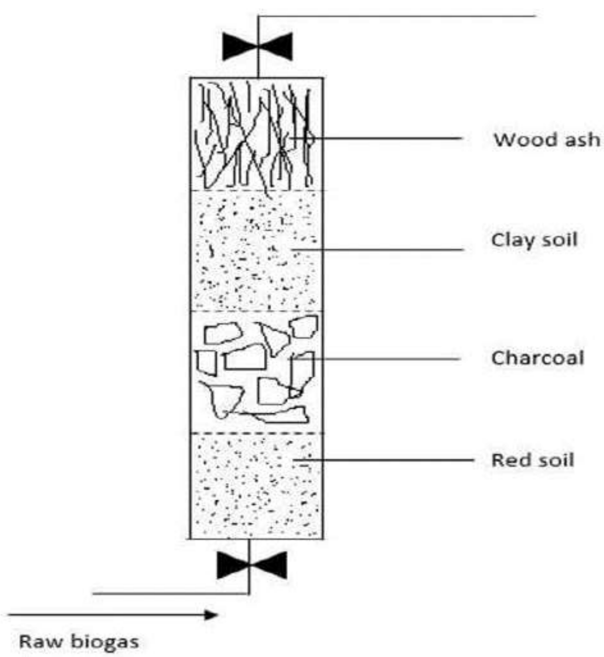

Figure 4: Array of combined adsorbent materials

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3.1 Adsorption Mechanism

Adsorption is a process in which molecules or atoms adhere to the surface of a solid or liquidmaterial (Kaithwas et al., 2012). The mechanism of adsorption is mainly affected by surface area, Van der Waals forces, chemical bonds, temperature and pressure.

The figure below depicts raw biogas purification through an ideal adsorption process.

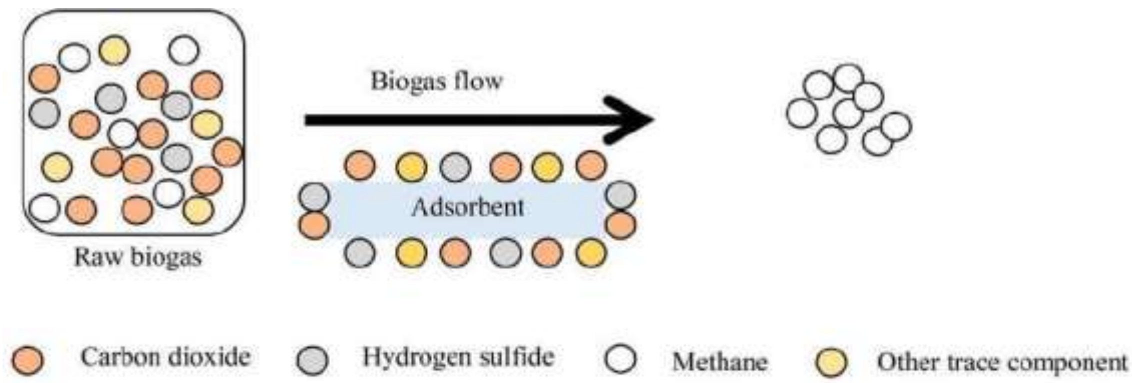

Figure 5: Adsorbent mechanism (Source: Mulu at al., (2021))

3.2 Bio-digester

The study relied on the existing bio-digesters at the IEET workshop, JKUAT, as the source of raw biogas that was used throughout the study period. The digesters are mainly fed with cow dung as the main feedstock. As is the case with most bio-digesters, each consists of an airtight,high-density polythene container within which pre-prepared cow dung is diluted in water flowcontinuously and is fermented by microorganisms present in the waste.

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Figure 6: A bio-digester at the IEET workshop, JKUAT

4.0 Results and Discussion

4.1 Results

From the study, raw biogas purification through adsorption using the low-cost adsorbent materials as highlighted earlier; gave the results as presented in the following table. All the readings were taken in triplicates and thus standard deviations were calculated and factored inthe data recordings.

Table 1: % methane after purification by individual materials

	Raw biogas	Red soil	Steel-wool	Charcoal	Iron shavings	Clay soil	Wood ash
a. %CH4	58.19±2.12	61.30±0.71	59.12±1.92	66.42±1.08	58.78±1.96	70.51±2.33	59.70±2.47
b. %CH4	56.03±2.12	61.21±0.71	56.55±1.92	68.73±1.08	56.06±1.96	64.84±2.33	58.39±2.47
c. %CH4	61.20±2.12	62.76±0.71	61.25±1.92	66.44±1.08	60.85±1.96	67.08±2.33	64.17±2.47

The study experiment was carried out in triplicates as shown above (a, b, c) and averaged asshown in the table below.

Table 2: Average % methane after purification by individual materials

	Raw biogas	Red soil	Steel-wool	Charcoal	Iron shavings	Clay soil	Wood ash

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Average % CH_4	58.47± 2.12	61.76± 0.71	58.97 ±1.92	67.20± 1.08	58.56± 1.96	67.48± 2.33	60.75± 2.47
Adsorbed impurities %	-	3.29	0.50	8.73	0.09	9.01	2.28

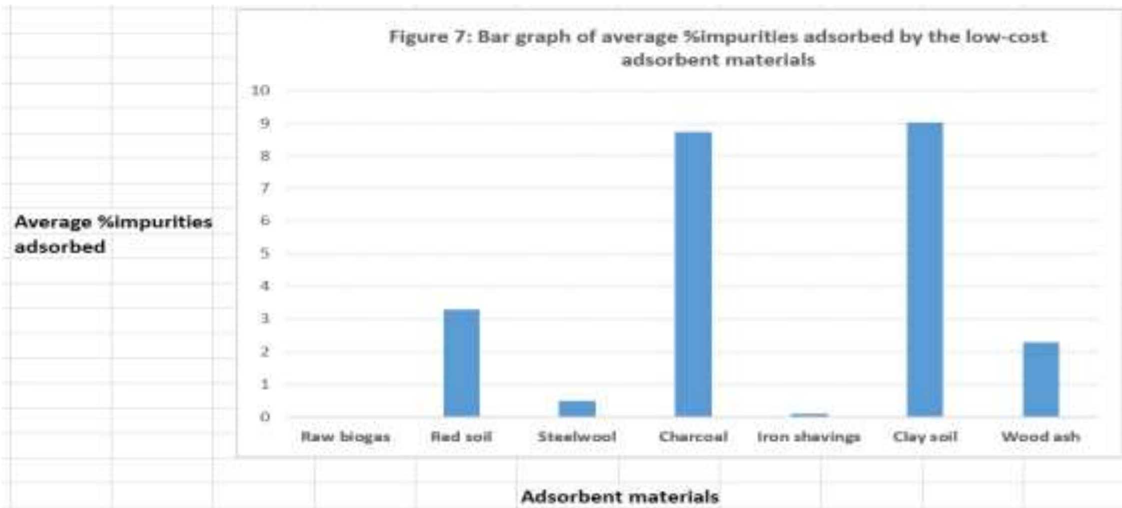

Figure 7: A bar graph of average %impurities adsorbed by individual adsorbent materials

Cleaning units of the adsorbent materials' physical interfaces/ combinations were also set up in twos, and also a combination of up to four materials as in figure 3. The biogas was analyzed in triplicates, both before and after passing through the materials; and resultant data recorded as in the table below.

Table 3: %methane after purification by combined adsorbent materials

Combinations	Raw biogas	Red soil and charcoal	Clay soil andwood ash	Red soil, charcoal, clay soiland wood ash
a) %CH_4	58.73±0.32	66.98±1.37	66.85±1.78	68.20±1.57
b) %CH_4	59.46±0.32	70.15±1.37	71.14±1.78	71.08±1.57
c) %CH_4	58.87±0.32	69.51±1.37	69.64±1.78	71.86±1.57

The above data was averaged and recorded as in the table below;

Table 4: Average %methane after purification by different adsorbents' combinations

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Combinations	Raw biogas	Red soil andcharcoal	Clay soil andwood ash	Red soil, charcoal,clay soil and wood ash
Average %methane	59.02±0.32	68.88±1.37	69.21±1.78	70.38±1.57
Adsorbed %impurities	-	9.86	10.19	11.36

The results in table 4 above were plotted on a bar graph as below;

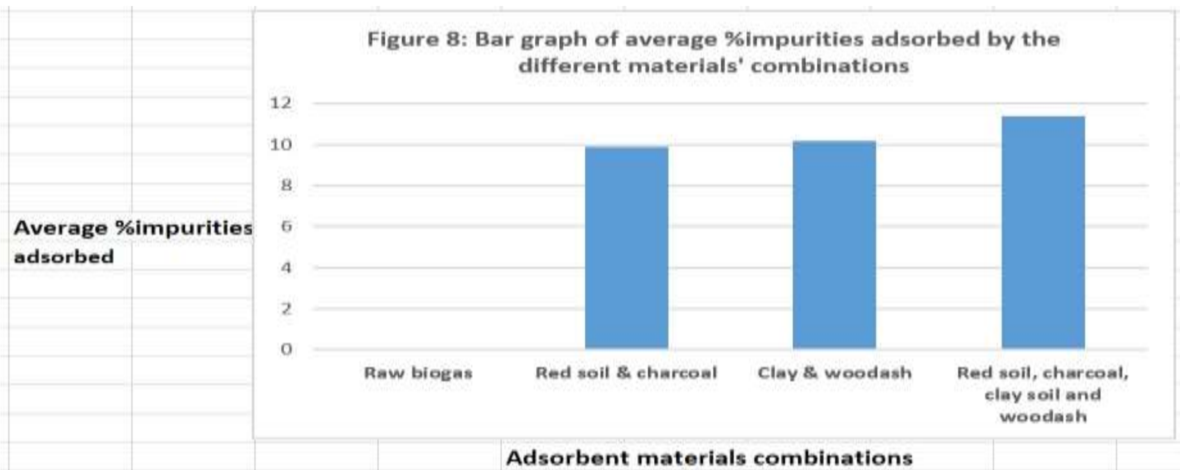

Figure 8: A bar graph of average percentage impurities adsorbed by the materials' combinations

4.2 Discussion of results

From the study's results, it was evident that the low-cost adsorbent materials, in their natural state, have a potential to purify biogas. Also notably clear, was that the different low-cost adsorbent materials exhibited different cleaning/adsorption capacities of the impurities present in raw biogas. Each of the adsorbent materials used was able to adsorb at least some impurities from the raw biogas, and this was evident in the improvement of the percentage composition levels of methane in the gas.

Purification by use of clay soil and charcoal recorded highest CH4 concentration of 67.48±2.33% (adsorption of 9.01%) and 67.20±1.08% (adsorptionof 8.73%) respectively, while steel-wool and iron shavings recorded the least methane concentration of 58.97±1.92% (adsorption of 0.50%) and 58.56±1.96% (adsorption of 0.09%) respectively. Use of steel-wool and iron-shavings thus recorded less that unit percentage of biogas impurities' adsorption. This can be attributed to the fact that the major biogas impurity; Hydrogen sulfide (H_2S), susceptible to adsorption by iron (Fe) is one of the trace components in raw biogas whose percentage levels in the gas is consequently low.

The relatively higher adsorption by clay soil could be attributed to its potential, due to its chemical

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composition, to adsorb carbon-dioxide from biogas both chemically and physically (Mulu et al., (2021)). Correspondingly, the adsorption performance by charcoal could be credited to its large surface area and porous nature that makes it suitable for adsorption of certain gases. Raw biogas purification by use of red soil and wood ash recorded methane concentration levels of 61.76±0.71% and 60.75±2.47% from 58.47±2.12% methane in raw biogas.

Further exploration of biogas purification by the low-cost adsorbent materials' combinations into one cleaning unit improved the purification to a percentage of 70.38±1.57% methane when red soil, charcoal, clay soil and wood ash were used; achieving an adsorption rate of 11.36%.

In Comparison with previous studies;

Ebunilo et al., (2016) in a study "Investigation of the purification of biogas from domestic wastes using local materials in Nigeria" compares well, from the resultant data, that natural adsorbents have a potential to purify biogas. In the study, Ebunilo et al., (2016) used natural adsorbents including; clay soil, charcoal, zeolite and iron ore haematite. Clay soil achieved adsorption of 2.08% improving %CH_4 to 71.13%, charcoal achieved an adsorption of 6.04 improving the methane levels to 74.89 while iron ore improved the CH_4 percentage to 69.68% by an adsorption of 0.60%. Combining all the adsorbents together, the study achieved an adsorption of 13.41% purifying the biogas to 82.37%. These results, despite the variations, compare well with those achieved in this study.

Other studies done before that agree with this study include;

i. A review by Mulu et al., (2019) on "A review of recent developments in application oflow cost natural materials in purification and upgrade of biogas."

ii. Muche et al., (1985) "The purification of biogas"

5.0 Conclusion and recommendation

Despite the disparities, the results showed that low-cost adsorbent materials have a potential in purification and upgrading of biogas

Purification of biogas can be effectively done using the existing commercial technologies. However, given their high initial investment cost and sustainability, an alternative biogas purification method should be sought. Low-cost adsorbent materials have the potential to provide this option, as depicted by the study results.

This study recommends further exploration of low-cost natural adsorbents as potential alternative method of biogas purification.

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6.0 Conflict of Interest

None.

7.0 References

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