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Enhancing biogas quality: An experimental investigation of CO_2 and H_2S removal techniques

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Abstract

The increasing demand for sustainable energy sources has brought biogas into focus as a promising alternative to fossil fuels. However, impurities like carbon dioxide (CO_2) and hydrogen sulfide (H_2S) in biogas pose challenges to its effective utilization. This study aimed to design a purification system to decrease these impurities, evaluating the efficacy of conventional methods (iron sponge and water scrubbing) and alternative approaches (activated carbon and NaOH solution) in removing CO_2 and H_2S from biogas. The experimental setup involved integrating the purification system with an anaerobic digester producing raw biogas from swine manure. The concentrations of CH_4, CO_2, and H_2S were measured before and after passing through the purification chambers. Although no H_2S was detected in the raw biogas, the study focused on CO_2 removal. Results showed that the activated carbon scrubber significantly reduced CO_2 levels from 35% to 20% after a 60-minute retention time. The NaOH solution demonstrated excellent CO_2 removal efficiency of 86.27%, while the water scrubber achieved only 8.98% efficiency. Both the iron sponge and water scrubber modestly increased the CH_4 concentration by 1.9% and 1.7%, after 15 minutes. However, the NaOH solution yielded a more substantial 6.6% increase in CH_4 concentration. This study highlights the potential of alternative methods like activated carbon and NaOH solution for effective biogas purification, particularly in removing CO_2 impurities and enriching the methane content, promoting sustainable energy solutions.

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Introduction

The increasing global demand for sustainable and renewable energy sources has brought biogas into the spotlight as a promising alternative to fossil fuels. Biogas, a combustible mixture of gases produced by the anaerobic digestion by the breakdown of organic matter, offers a clean and environmentally friendly energy solution (Rajendran *et al.*, 2012). It is primarily composed of methane (CH_4) and carbon dioxide (CO_2) with smaller amounts of hydrogen sulfide (H_2S), ammonia (NH_3) and nitrogen (N). However, the presence of impurities, particularly carbon dioxide and hydrogen sulfide, poses significant challenges to the effective utilization of biogas (Abatzoglou and Boivin, 2009). Carbon Dioxide is a biogas impurity with 30-40% of the biogas composition which reduces the calorific value and burning velocity of biogas leading to incomplete combustion and decreased engine efficiency (Bari, 1996). On the other hand, Hydrogen Sulfide (H_2S) is a toxic and corrosive gas. Exposure to H_2S can cause severe health hazards and damage equipment (Dorman, 2010). The toxic effects of hydrogen sulfide are dose-dependent, affecting the nervous, cardiovascular, and respiratory systems. Individuals exposed to approximately 100 ppm of hydrogen sulfide may experience symptoms such as nausea, vomiting, diarrhea, and cardiac arrhythmias (Dorman, 2010). Additionally, H_2S must be eliminated from biogas produced by anaerobic digestion systems to reduce the engine-generator set maintenance expenses (Choudhury *et al.*, 2019).

Numerous purification techniques have been explored to purify biogas by removing CO_2 and H_2S. While conventional methods, like water scrubbing and chemical absorption, have been widely adopted due to their effectiveness and relatively low cost (Bauer *et al.*, 2013; Tock *et al.*, 2010), they require substantial infrastructure and less suitable for small-scale biogas production systems (Yousef *et al.*, 2016). Alternative approaches, including adsorption processes using activated carbon and iron-based materials have gained attention for their potential to remove H_2S and other impurities efficiently (Sithole *et al.*, 2017; Xu *et al.*, 2002). Cristiano *et al.* (2020), highlight iron oxide nanoparticles as a simple desulfurization method with potential for adsorbent regeneration. These nanoparticles offer improved H_2S removal efficiency and longer adsorbent lifespan, minimizing waste. Additionally, low-temperature, high pressure conditions can enhance absorption rates (Deng *et al.*, 2020). Chemical absorption, involving a solvent interaction with CO_2, further contributes to biogas quality improvement (Xu *et al.*, 2020). In the Philippines, where many rural and remote areas still lack access to traditional energy sources, a study aims to design a purification system that will decrease the biogas impurities of CO_2 and H_2S. By evaluating the efficacy of both conventional and alternative methods for biogas purification, this study seeks to identify the most effective strategies for removing CO_2 and H_2S present in biogas. The conventional method employs reagents such as iron sponge and water, while the alternative approach utilizes activated carbon and NaOH (sodium hydroxide) solution. Activated carbon and iron sponge are specifically targeted at removing H_2S, whereas NaOH solution and water scrubbers are employed for CO_2 removal (Awe *et al.*, 2017; Tippayawong and Thanompongchart, 2010).

Materials and methods

Experimental set-up

The experimental procedure was designed to evaluate the purification system in removing CO_2 and H_2S impurities from raw biogas. The experimental setup for biogas purification involved integrating essential materials and components. The process began with raw biogas obtained from the anaerobic digestion of swine manure as the input. The swine manure was weighed and mixed with water at a ratio of 1:1 to prepare the feedstock for the digester within 30 days retention time and stored in the floating drums for testing. The raw biogas produced from the digester was introduced into the purification system. Before and after passing through the purification scrubbers comprising activated carbon and NaOH solution, the concentrations of CH_4, CO_2, and H_2S in the raw biogas were measured using a portable gas analyzer.

Biogas purification system set-up

As shown in Fig. 1, a 200L airbed was used to collect raw biogas from a small-scale anaerobic digester, which was connected to the purification chambers. The chambers were thoroughly sealed and coated with epoxy resin to prevent leakage. Bubble testing ensured the integrity of the purification system before the experiment. The raw biogas was injected into the scrubber and retained for 15, 30, and 60 minutes, respectively.

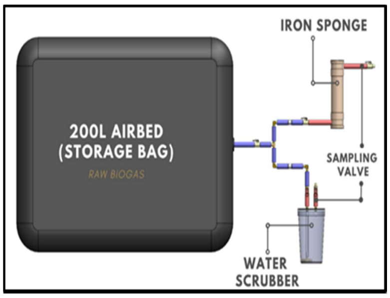

Fig. 1. Experimental diagram of the purification system setup

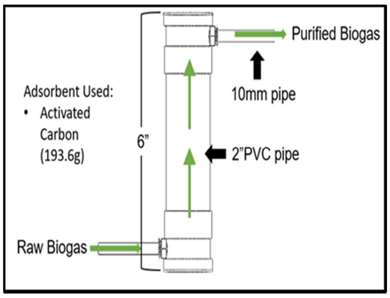

Fig. 2. Experimental setup for the H_2S removal System

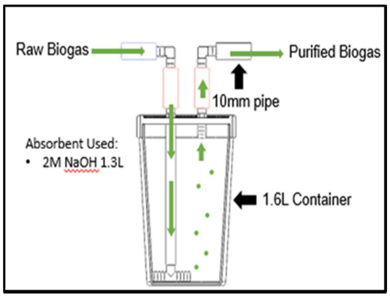

Fig. 3. Experimental setup for the CO_2 removal system

The purification system removed impurities from the biogas through adsorption and absorption processes. The input raw biogas flowed through the system containing agents like iron filings and water scrubbers. The adsorption process was used to remove H_2S using activated carbon and iron sponge. Water and NaOH solution, on the other hand, utilized the absorption process to remove CO_2.

For the conventional method, one chamber contained 76.3g of iron sponge, and another chamber contained a 1.3L water scrubber for H_2S and CO_2 removal. In the alternative method, one chamber was packed with 193.6g of activated carbon for H_2S adsorption, while a separate chamber housed a 1.3L NaOH solution with a concentration of 2M for CO_2 absorption (Fig. 2).

The CO_2 removal system as shown in Fig. 3 consisted of a 1.6L container holding 2M NaOH solution. Before introducing the raw biogas into the system, its composition was analyzed using a gas analyzer. The biogas was then pumped into the inlet pipe, which was submerged in the NaOH solution, allowing the gas to bubble through the liquid. This bubbling process significantly increased the surface area of contact between the biogas and NaOH solution, enhancing the reaction efficiency. As the biogas bubbled through the NaOH solution, the carbon dioxide present in the biogas reacted with the sodium hydroxide to form sodium carbonate, effectively removing CO_2 from the gas stream. The bubbles burst upon reaching the empty headspace above the liquid level within the container. This headspace was connected to an outlet pipe that led to a gas analyzer, which continuously monitored and measured the concentrations of methane (CH_4) and the remaining CO_2 in the purified biogas.

To evaluate the CO_2 removal performance, raw biogas was pumped into the scrubber's inlet pipe and stored within the chamber. The initial run had a biogas retention period of 15 minutes inside the chamber, during which the composition of the purified gas was tested at 15-minute intervals. This experimental run was repeated four times, resulting in a total experimental duration of 60 minutes. The systematic bubbling process, coupled with the chemical reaction between CO_2 and NaOH, aimed to achieve efficient

CO_2 removal from the raw biogas, and enhancing the quality and purity of the biogas for power generation applications.

Results and discussion

The concentrations of CH_4, CO_2, and H_2S in the raw biogas were analyzed using a gas analyzer. Unfortunately, no H_2S was detected in the raw biogas used in the experiment, as it showed 0 ppm on the device. Consequently, CO_2 became the primary focus of the investigation. The absence of H_2S in the biogas can be attributed to the specific characteristics of the feedstock (swine manure) and the anaerobic digestion conditions employed. Previous studies have reported that swine manure typically generates low levels of sulfur compounds during anaerobic digestion, especially when the process operates under favorable conditions with sufficient alkalinity and balanced nutrient levels (Rashed *et al.*, 2019; Shen *et al.*, 2021).

H_2S removal

Although no H_2S was detected, the scrubber containing 76.3g of iron filings was still utilized to evaluate its potential for removing other impurities present in the biogas. The raw biogas was fed and stored into a chamber containing the iron sponge in the form of iron filings. Initially, the raw biogas was held for a retention time of 15 minutes, allowing the adsorbent material to interact and purify the raw biogas. The modest reduction in CO_2 concentration observed in this study from 35% to 33% shows that the iron filings may possess some adsorptive capacity for CO_2. This potential adsorption could be attributed to the formation of iron oxides and oxyhydroxides on the surface of the filings, which can act as active sites for CO_2 adsorption (Bhargava *et al.*, 2022; Spigarelli and Kawatra, 2013). However, it is crucial to note that the low surface area and porosity of the iron filings compared to dedicated CO_2 adsorbents like activated carbon limit their overall adsorption capacity.

CO_2 removal

The scrubber containing activated carbon was also utilized to investigate its effect on CO_2 removal and other potential impurities. Activated carbon is widely recognized for its adsorptive properties and has been extensively studied for biogas upgrading applications (Kadam and Panwar, 2017; Xu *et al.*, 2020). The results obtained from the activated carbon scrubber shows a significant reduction in CO_2 concentration, with levels decreasing from 35% to 28% after a 15-minute retention time (Fig. 4). Extending the retention time to 30 and 60 minutes further decreased the CO_2 levels to 24% and 20%, respectively (Fig. 5). These findings align with the well-established principles of adsorption, where longer residence times and increased contact between the adsorbate (CO_2) and adsorbent (activated carbon) facilitate enhanced mass transfer and higher removal rates (Koubaissy *et al.*, 2019; Zhang *et al.*, 2021).

The high surface area and porous structure of activated carbon provide a favorable environment for CO_2 adsorption, making it an effective adsorbent for biogas upgrading.

The water scrubber used in this study only removed a small amount of CO_2, with an efficiency of 8.98% as shown in Fig. 6. This finding agrees with previous study stated that CO_2 does not dissolve well in water, making water scrubbing not very effective for significant CO_2 removal (Islamiyah *et al.*, 2014; Tobiesen *et al.*, 2018). The CO_2 removal efficiency of the water scrubber during its initial 15-minute operation was 21.2%, differences in experimental setup and operating conditions could explain the variation compared to our study (Gao *et al.*, 2020). Additionally, factors like temperature, pressure, and other dissolved substances can impact how well CO_2 dissolves in water (Xu *et al.*, 2020), potentially contributing to the different removal efficiencies observed.

The activated carbon (AC) used in this study did not effectively remove CO_2, although it requires pre-treatment to be effectively used for CO_2 removal. Many studies highlight that such modifications improve AC's ability to adsorb and selectively capture CO_2 (Peredo-Mancilla *et al.*, 2019; Rattanaphan *et al.*, 2019; Yahya *et al.*, 2015; Rashidi and Yusup, 2017).

In contrast, the 2M sodium hydroxide (NaOH) solution showed excellent CO_2 removal efficiency of 86.27% from an initial concentration of 16.82%. This finding aligns with the study by Maile *et al.* (2017) stated, the increasing of NaOH concentrations, resulted in greater and higher removal efficiency.

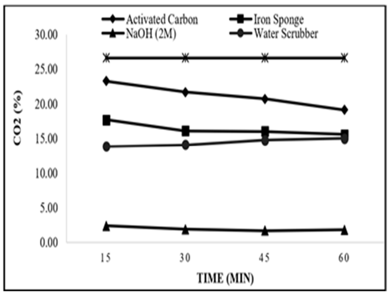

Fig. 4. Purified biogas CO_2 content 15 minutes retention time

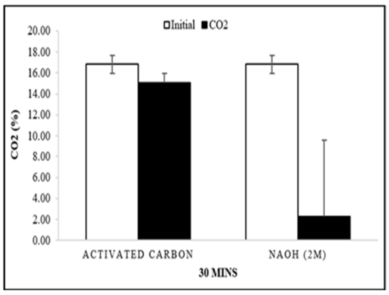

Fig. 5. Purified Biogas CO_2 Content 30 minutes retention time

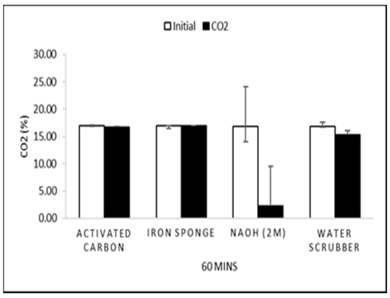

Fig. 6. Purified Biogas CO_2 Content 60 minutes' retention time

increase of approximately 6.6% in the CH_4 concentration. This finding aligns with the study by Xu *et al.* (2019), who reported that NaOH solution effectively removes CO_2 and H_2S from biogas, thereby increasing the CH_4 concentration and enhancing the methane concentration in biogas.

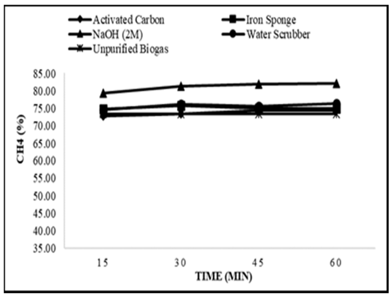

Fig. 7. Purified Biogas CH_4 Content 15 minutes' retention time

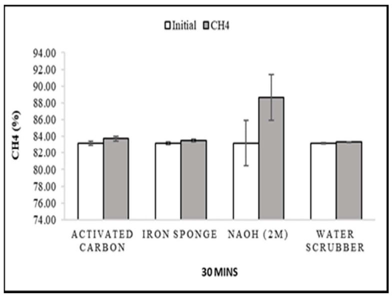

Fig. 8. Purified Biogas CH_4 Content 30 minutes' retention time

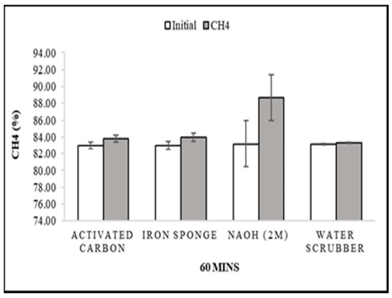

Fig. 9. Purified Biogas CH_4 Content 60 minutes' retention time

Methane

The concentration of CH_4 increased after passing through the purification system and enriching the methane content. The activated carbon had a minimal impact; it still contributed to the overall methane enrichment process as shown in Fig. 7. The use of NaOH solution yielded promising results, with an

The comparison between the water scrubber and the iron sponge reveals that both techniques effectively improved the CH_4 concentration, the results indicate a 1.7% and 1.9% increase in CH_4 concentration, from the baseline of 73.39% unpurified biogas after a 15-minute retention time. This finding suggests that the adsorption and absorption media utilized were comparable in terms of efficacy, aligns with the study by Khunprasert and Charinpanitkul (2022), who reported similar performances of water scrubbers and iron sponges in biogas purification. However, the second and third runs demonstrated that the water scrubber's efficacy in increasing the CH_4 concentration was lower compared to the iron sponge, even with extended retention times, as shown in Fig. 8 and 9. This observation could be attributed to the saturation of the water scrubber over time, as stated by Rajendran *et al.* (2020), who noted that water scrubbers may become less effective due to the accumulation of dissolved impurities.

As shown in Fig. 8 for the 30-minute retention time, the iron sponge demonstrated a higher percentage increased from an initial value of 83.18% to 83.54%, compared to the water scrubber, which only grew by approximately 0.43%. This finding suggests that the iron sponge may be more effective in purifying biogas and increasing the CH_4 concentration, particularly at longer retention times. However, it is crucial to consider that both reagents had almost the same amount of CH_4 increases, indicating that their overall performance was comparable. This observation aligns with the study by Aghbashlo *et al.* (2021), which reported that iron sponges outperformed water scrubbers in biogas upgrading, particularly at higher operating times.

Comparing the results with the study by Maile *et al.* (2017) employed a 1-3M NaOH solution contained in a 500 mL vessel, through which biogas was bubbled. In the researcher's experiment, a similar procedure was carried out using a 1.6L container containing a 2M NaOH solution. Maile *et al.* reported a significant increase in CH_4 concentration by 43%, from 52% to 74%, after passing the biogas through the 2M NaOH solution. However, 6.6% increase in CH_4 was observed, this discrepancy in the CH_4 concentration increases the initial low CO_2 concentration in the biogas feedstock used in the experiment. Acharya *et al.* (2021) highlighted that the effectiveness of NaOH solution in biogas upgrading depends on the initial CO_2 concentration, with higher CO_2 levels leading to more significant CH_4 enrichment. And the effectiveness of NaOH in increasing the CH_4 concentration is primarily due to its ability to remove CO_2 through chemical absorption. If the initial CO_2 concentration is low, the potential for increasing the CH_4 concentration may be limited (Andriani *et al.*, 2014).

Conclusion

The iron sponge showed limited effectiveness in removing sulfur compounds due to the lack of significant H_2S in the raw biogas, as detected by the gas analyzer. However, it removed a small amount of CO_2, with 2.14% removal efficiency, resulting in a minor increase in CH_4 concentration. And the NaOH solution outperformed activated carbon in removing CO_2, as activated carbon primarily targets the removal of H_2S. Even with longer retention time, activated carbon did not substantially improve the CH_4 concentration due to its ineffectiveness in CO_2 removal.

Recommendation(s)

It is recommended to use NaOH solution concentrations between 2M and 3M, as higher concentrations have been proven effective in CO_2 removal without compromising the removal efficiency. And to evaluate the maximum performance capability of the purification agents, it is advisable to use a raw biogas feedstock with significant concentrations of impurities. The researcher in the present study used raw biogas with minimal impurities, which may have limited the purification agent's ability to remove substantial amounts of impurities. By implementing these recommendations, future researchers can potentially optimize the biogas upgrading process, achieve higher CH_4 concentrations, and contribute to the advancement of sustainable energy solutions.

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