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The Optimization of Biogas Upgrading: The CO_2 Adsorption on Sugarcane Bagasse-Based Biochar and Zeolite

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Most households in Indonesia use liquefied petroleum gas (LPG) for cooking. Still, the increasing import of LPG is feared to burden Indonesia's current trade balance. Therefore, to meet the demand, it needs other alternative energies. Biogas is a promising alternative energy source for households because it can be produced from agricultural waste or animal manure. Nevertheless, sustainable biogas adoption faces many challenges, including the presence of carbon dioxide (CO_2), which affects the calorific value of biogas. The removal of CO_2 from biogas is known as biogas purification and is often carried out using carbon material (charcoal) as a CO_2 adsorbent. Carbon-based biomass waste as a CO_2 adsorbent in biogas is an uncommon practice. So far, biomass waste has usually been investigated for biogas upgrading on a laboratory scale using synthetic biogas. This study investigated the use of biomass waste, sugarcane bagasse-based biochar, and natural zeolite, in the different adsorption contact times for increasing the calorific value of biogas by CO_2 adsorption. The CO_2 adsorption was carried out in a biogas purificator in adsorption times of 10, 15, 20, and 25 minutes at room temperature and gas pressure ranging from 6 to 8 bar. The CO_2 adsorption capacity was further reduced by increasing the adsorption time. The combination of sugarcane bagasse-based biochar and zeolite exhibited a high CO_2 removal capacity of 78.49% at 10 minutes of adsorption time. The increase in adsorption time saturated the CO_2 adsorption capacity. The lowest CO_2 adsorption capacity was achieved at 25 minutes of adsorption time with a CO_2 removal capacity of 12.34%.

Keywords: adsorption, biochar, biogas, CO_2, sugarcane-bagasse.

Introduction

According to the Indonesia Energy Outlook (Secretary General National Energy Council, 2019), Indonesia's biomass energy capacity from animal and plant (edible and inedible) raw materials is 32,654 MW. These resources can be converted into biogas energy, which can help meet the nation's energy demand and serve as a substitute for liquefied petroleum gas (LPG). In addition, biomass resources can also be utilized through other conversion pathways such as direct combustion, pyrolysis, and gasification, broadening their role as renewable energy sources. Since the conversion program from kerosene to LPG, LPG consumption has increased. According to the Center for Assessment of Process Industry and Energy (2020), the demand for LPG is projected to grow from 7.2 million tonnes in 2017 to 17.4 million tonnes in 2050. In contrast, in 2024, LPG production was only 2 million tonnes. Indonesia is concerned that the growing import of LPG may negatively impact its trade balance. Therefore, it is necessary to explore other energy sources to meet the increasing energy demand. Consequently, alternative energy like biogas for household cooking needs to be encouraged. Biogas serves as a source of cooking energy and is also utilized as a fuel in transportation, industry, and the commercial sector. Biogas is a significant contributor to the shift towards low-carbon energy. The biogas production pathway, from biomass to biogas, involves the anaerobic digestion of organic waste or manure. Biogas is one of many types of renewable energy fuel that can help mitigate greenhouse gas emissions, reduce deforestation, and improve environmental health (Cuellar and Webber, 2008). The energy generated by biogas is more sustainable and renewable, which can contribute to an endless stream of socioeconomic benefits (Geddafa et al., 2023). A biogas plant from cattle manure, suitable for a household, would effectively reduce greenhouse gas emissions that would otherwise be released into the atmosphere through the combustion of LPG, kerosene, and traditional open-air burning of firewood (Pradhan and Limmeechokchai, 2017), while biogas slurry, a residual product of biogas generation, still can be utilized as a replacement for chemical fertilizer. Replacing chemical fertilizer with biogas slurry decreases the release of carbon dioxide (CO_2) and nitrous oxide. Carbon dioxide, or CO_2, is a greenhouse gas that causes global warming. According to Pathak et al. (2009), a biogas plant designed for a household with 4 cattle, which generates 4400 kg of dung, can create 2200 m^3 of biogas per year. According to Pathak et al. (2009), biogas saves global warming potential (GWP) from combusting kerosene and firewood for cooking or lighting by 762 and 10,571 CO_2 equivalents, respectively, and the GWP from chemical fertilizer by 302 CO_2 equivalents. Pertiwiningrum et al. (2020) also revealed that biogas emits less CO_2 emission and CO_2 flux between cattle's buried manure and fresh manure, compost, and biogas sludge by comparing methane (CH_4). This means biogas is revealed to be more environmentally friendly in biomass waste management.

In Indonesia, the cattle population is growing rapidly. In 2014, the cattle population was 14.7 million heads and grew positively to 16.9 million in 2019 and 2020, reaching 17.4 million (Indonesia Central Bureau of Statistics, 2021). Population growth, economic development, and changes in consumer preferences are leading factors that have become driving forces for livestock industries in Indonesia. Beef consumption in Indonesia was around 2.4 kg per capita per year in 2023 and is predicted to increase to double or triple in the coming years as urbanization and household income increase (Center for Agricultural Data and Information Systems, 2023). The other fact is that cattle in Indonesia mostly use a smallholder farming system. Generally, they do not treat waste properly; some even pile up the cattle manure. The cattle manure has the potential to be converted to energy by biogas technology. Biogas must be improved by CO_2 removal technology to be adopted by households and used sustainably. Budzianowski (2016) defined adsorption as a technique that is used for CO_2 removal in biogas, among other approaches. By adsorption, biogas produces high methane and calorific value. Dutta and Mahanta (2014) explained that natural zeolite and clay are nature-based materials used as CO_2 adsorbents. In addition, activated carbon, biochar, and flying ash from biomass waste have been widely developed as alternative adsorbents.

Besides livestock manure, sugarcane industry waste is a significant and potential source of biomass in Indonesia that can be utilized for biogas production. Sugarcane businesses are agricultural enterprises that generate substantial amounts of waste as a byproduct of sugar production. As of 2016, Indonesia had a total of 63 sugar mills, which were controlled by 18 different businesses. Toharisman and Triantarti (2016) reported that these mills had a combined running capacity of 245,900 tonnes of cane per day, and in 2019, Indonesia had a total operational capacity of 278,411 tonnes of sugarcane per day (McDonald and Meylinah, 2019). It means that waste from sugary industries will increase with the increase in sugar production capacity in Indonesia, and this waste can be used as new raw materials, energy, or other applications. The first step in sugar production from sugarcane is juice extraction or cane crushing using crushing rollers. After juice extraction, the fibrous material is known as bagasse (Bhatnagar et al., 2016). Bhatnagar et al. (2016) reported that the bagasse yield for every 1000 tonnes of sugarcane is 27%. Meghana and Shastri (2020) found that the conversion of one tonne of sugarcane results in an output of 0.3 tonnes of bagasse. The bagasse consists of around 26–47% cellulose, 19–33% hemicellulose, 14–23% lignin, and a tiny proportion of other substances (Mahmud and Anannya, 2021). The utilization of bagasse usually produces energy, paper, and cattle feed (Bhatnagar et al., 2016). Lignocellulosic feedstock in bagasse can also be used for energy (Meghana and Shastri, 2020) or other applications.

Sugarcane bagasse is also used in many applications, such as an adsorbent by a carbonization process. In biogas applications, sugarcane bagasse is used as a raw material of biogas and can also be used as an adsorbent for biogas purification. The conversion of waste from sugary industries to energy or other applications is revealed to reduce CO_2 emissions (Mohammadi et al., 2020). Using biomass waste as raw material for bioenergy production and other applications would contribute to circular economy practice (United Nations Development Programme, 2021). This conversion leads to implementing the circular economy concept for sustainable management in sugary industries. A circular economy is defined as generating economic growth by the value of the resource as long as possible by reducing, reusing, recycling, refurbishing, and renewing or recovering (see *Table 1*). Sugarcane bagasse can be re-utilized as a precursor of biogas adsorbent, functioning as a renewable fuel in sugary industries, thereby contributing to a reduction in production costs. Converting bagasse to biochar has many potential applications, like pollutant remediation, CO_2 adsorption, an additive in composites, and a catalyst in biofuel production (Zafeer et al., 2024).

Table 1. *The Circular economy approach by the 5Rs practice (United Nations Development Programme, 2021)*

Items	Definition
Reduce	Remove waste in the production and supply chain. Reduce energy usage. Virtualize products and services. Redesign the product to require fewer resource inputs.
Reuse	Collaborate on resources. Utilization of pre-owned goods. Enhance the usefulness of assets by providing them in the form of services.
Recycle	Reuse resources. Utilizing anaerobic digestion to extract biochemicals from organic waste.
Refurbish	Recondition items or components. Extend the lifespan of the product with regular maintenance.
Renew	Give priority to the use of renewable energy sources and materials.

Biogas purification is the process of purifying biogas that can reduce combustion capacity by CO_2 adsorption. Numerous factors influence the performance of biogas purification. Variations in the physical characteristics of adsorbents influence adsorption performance (Ketabchi et al., 2023). The CO_2 adsorption undergoes physical and chemical adsorption at the surface of the adsorbent. The adsorbent's basicity and pore size are key parameters that affect CO_2 adsorption (Mfoumou et al., 2018). Besides internal factors, external factors like temperature, air pressure, and contact time also influence CO_2 adsorption. According to Boonpoke et al. (2011), for the temperature range of 30–150°C, The CO_2 adsorption capacity on activated carbon decreased as adsorption (Lee and Park, 2014) revealed that the 5–7 bar range is the ideal pressure for CO_2 adsorption. For contact time, Creamer et al. (2014) observed that the kinetic CO_2 adsorption process occurred very quickly in the first ten minutes, slowed down after ten minutes, and flattened at 60 minutes. This means that the equilibrium was reached within 60 minutes of contact time. The same phenomenon was also reported by Hauchhum and Mahanta (2014) and Xu et al. (2016).

This study investigated the influence of adsorption contact time on biochar-based sugarcane bagasse. The investigation was conducted to find the optimal contact time in biogas purification. Hopefully, the improvement in biogas purification technology will increase the adoption of renewable energy in Indonesia.

Methods

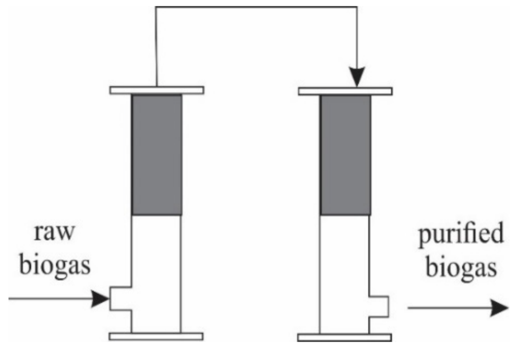
The sugarcane bagasse obtained from the surrounding area was still moist and had to be dried in the sun for a few days. Next, the sugarcane bagasse underwent pyrolysis at a temperature of 255°C for 3 hours following the method of Nguyen and Lee (2016). Sugarcane bagasse-derived biochar was employed as a CO_2 adsorbent in the process of purifying biogas. The adsorption of CO_2 was performed at ambient temperature and a pressure range of 5–7 bar (Lee and Park, 2014). The adsorption column employed in this work is a packed bed column of the second level, measuring 20 cm in length and 3 cm in diameter (*Fig. 1*). Each column was filled with an adsorbent with the formulation specified in *Table 2*. The column was immediately linked to the biodigester and biogas holder. An air compressor moved the biogas mixture through the column to facilitate the process of adsorbing CO_2. The most effective adsorbent for reducing CO_2 was employed in the treatment process with varying contact times. The contact time was varied for durations of 10, 15, 20, and 25 minutes. Biogas samples were identified for the CO_2 composition level by gas chromatography. The reduction of the CO_2 level after adsorption can be calculated with the formula, using *Eq. 1*:

$$\% \, CO2 \; reduction = \frac{CO2 \; before \; adsorption - CO2 \; after \; adsorption}{CO2 \; before \; adsorption} \quad (1)$$

Table 2. *Formulation of adsorbents for CO_2 adsorption in biogas*

Items	Adsorbents (%volume)	
	Biochar	Zeolite
ZZ (Zeolit)	0	100
ZB1 (Zeolit-Biochar)	25	75
ZB2 (Zeolit-Biochar)	50	50
ZB3 (Zeolit-Biochar)	75	25
ZB4 (Zeolit-Biochar)	100	0

Fig. 1. *The CO_2 adsorption column*



The calorific value test of biogas was conducted in this study to identify the calorific value of biogas as a renewable fuel. The purified biogas was packaged in a bottle tank with a capacity of 4 bars and used for heating one liter of water until the biogas was empty. The calorific value was calculated with the formula using *eq. 2*:

$$Q = m \, x \, Cp \, x \, (T_f - T_i) \quad (2)$$

The *Q* refers to the calorific value of biogas (kJ), *m* refers to the mass of 1 liter of water, *Cp* refers to the water-specific calorific value (kJ/°C), T_f and T_i refer to the initial and finish temperature of water. The calorific values were then investigated for the correlation with the CO_2 reduction in each treatment. Combustion efficiency was also calculated in this study to identify the efficiency of biogas production by comparing it to the theoretical calorific value of biogas. The theoretical calorific value of biogas was determined by multiplying the mole fraction of methane in biogas by the specific calorific value of one mole of methane.

Results and Discussion

The concentration of CO_2 as an impurity gas in biogas is a crucial parameter for assessing the biogas quality. A decrease in CO_2 levels indicates a higher grade of biogas. The findings of the CO_2-induced changes in composition (*Table 3*) served as a point of reference for the experiment investigating the effects of varying contact times.

Table 3. *The decrease in CO_2 at different adsorbent compositions*

Items	Before adsorption	After adsorption	The reduction of CO_2
	ppm	ppm	%
ZZ (Zeolit)	214,700	229,000	−6.00
ZB1 (Zeolit-Biochar)	261,360	213,540	17.33
ZB2 (Zeolit-Biochar)	178,720	38,445	78.49
ZB3 (Zeolit-Biochar)	221,020	203,720	7.83
ZB4 (Zeolit-Biochar)	180,830	212,020	−17.25

Every volume adsorption column has been filled with adsorbents. The 100% volume of zeolite (referred to as ZZ) means that each column contained the same amount of zeolite as the volume dimension of each adsorption column. The differences in the volume composition of adsorbents in each adsorption column affected the ability of CO_2 capture in biogas, and it has been mentioned in *Table 3*. The results showed that the adsorbent composition consisting of 50% volume of sugarcane bagasse-based biochar and 50% volume of natural zeolite (ZB2) achieved the greatest reduction in CO_2 levels, with a decrease of 78.49%. The treatment using 100% sugarcane bagasse-based biochar (ZB4) showed the smallest decline, with a reduction of −17.25%. In this study, the use of zeolite did not positively influence the reduction of CO_2. Based on our previous study (Wuri et al., 2021). The CO_2 reduction was caused by the zeolite's pore radius being bigger than biochar. However, zeolite has a larger surface area. The larger pore radius area made CO_2 molecules pass the adsorbents so that there was no reduction of CO_2 instead of an increase. The increase in CO_2 may have been caused by the air addition that is input during the adsorption process using the air compressor. There was a slight drop in CO_2 when the volume of biochar increased beyond 50%, but after the volume reached 100%, there was an increase in CO_2. The substitution of zeolite with sugarcane bagasse-based biochar increases the opportunity for CO_2 adsorption because, based on the pore characteristics tests conducted in our previous study (Wuri et al., 2021). The biochar exhibits a greater volume of micropores than zeolite. According to the findings presented in *Table 3*, the mixture containing equal volumes of zeolite and biochar (referred to as ZB2) exhibited the highest CO_2 adsorption capacity, measuring 78.49%. A balanced combination of zeolite and biochar will maximize CO_2 adsorption because they have different properties in their absorption but complement each other. Ritonga and Masrukhi (2017) stated that zeolite is hydrophilic and polar, binding oxygen. At the same time, biochar is hydrophobic and nonpolar, which can bind carbon, so combining the two absorbents will maximize absorption. Boer et al. (2023) claimed that to enhance CO_2 adsorption, natural zeolites and biomass-based absorbents need to be further activated to have bigger pores.

In this study, the combination of 50% volume of zeolite and 50% biochar (ZB2) was used for the next experiment in the variation of contact times of 10, 15, 20, and 25 minutes. Many studies (Alonso-Vicaro et al., 2010; Hauchhum and Mahanta, 2014; Creamer et al., 2014) have reported CO_2 adsorption using adsorbent-based biomass resources. However, there has been a lack of studies focusing on how variations in the duration of adsorption contact time influence the adsorption capacity (*Table 4*).

Table 4. *The studies of CO_2 adsorption*

Reference	CO_2 adsorbent	Gas	Composition Test	Results
Boonpoke et al. (2011)	Activated carbon from rice husk Activated carbon from bagasse	Pure CO_2	Thermogravimetric Analysis (TGA)	CO_2 adsorption capacity from bagasse higher than rice husk The high adsorption capacity to its surface area, high carbon content, and low ash
Hauchhum and Mahanta (2014)	Activated carbon from coconut fiber Zeolite	CO_2 and air	Flue Gas Analyzer (FGA)	The best CO_2 adsorption performed by zeolite 13X
Creamer et al. (2014)	Biochar from hickory wood Biochar from sugarcane bagasse	Pure CO_2	Thermogravimetric Analysis (TGA)	The best CO_2 adsorption was performed by biochar from sugarcane bagasse Findings from this work showed that the CO_2 capture ability of most tested biochar samples also increased with the surface area.

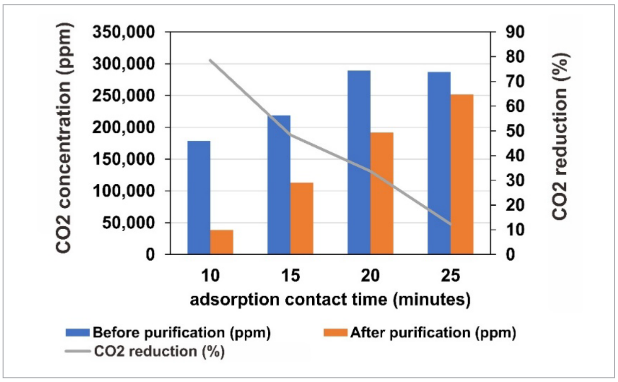
Reference	CO_2 adsorbent	Gas	Composition Test	Results
This study	Combination of zeolite and biochar from sugarcane bagasse	Biogas	Gas chromatography	This study investigated the use of biomass waste, sugarcane bagasse-based biochar, and natural zeolite with a volume ratio of 50%, and 50% in the different adsorption contact times for increasing the calorific value of biogas by CO_2 adsorption. The CO_2 adsorption was carried out in a biogas purificator in adsorption times: 10, 15, 20, and 25 minutes at room temperature and gas pressure ranging from 6 to 8 bar. The CO_2 adsorption capacity was further reduced by increasing adsorption time. The combination of sugarcane bagasse-based biochar and zeolite exhibited a high CO_2 removal capacity of 78.49% at 10 minutes of adsorption time. The increase in adsorption time saturated the CO_2 adsorption capacity. The lowest CO_2 adsorption capacity was performed at 25 minutes of adsorption time with a CO_2 removal capacity of 12.34%.

A previous study about the impact of adsorption contact time on the adsorption capacity of sugarcane bagasse biochar has been investigated by Creamer et al. (2014). Creamer et al. (2014) reported that CO_2 adsorption was very fast at the beginning, slowed down after about 10 minutes, and finally reached a plateau after 60 minutes, indicating it was approaching equilibrium. The experiment was conducted using pure CO_2 gas. However, the CO_2 adsorption experiments using biogas (mixing gases) are infrequently reported. Therefore, the objective of this study is to investigate CO_2 adsorption using biogas and determine the most effective parameters for purifying biogas using biochar derived from sugarcane bagasse.

So our study utilized the maximum CO_2 adsorption capacity identified in the prior study as the basis for the subsequent experiment, focusing specifically on the variation in contact time. Gas samples collected both before and after adsorption were sent for gas chromatography analysis following the experiment. The results are shown in *Fig. 2*.

The results in *Fig. 2* demonstrate that changes in contact times have an impact on the reduction of CO_2. The adsorption of CO_2 first increased during the purification process and subsequently decelerated after ten minutes. The most effective CO_2 adsorption was achieved by using a mixture of 50% zeolite and 50% biochar (ZB2) with an adsorption contact period of 10 minutes. It can be inferred that as the duration of adsorption increases, the concentration of CO_2 decreases. It occurred due to the maximal adsorption capacity in the first ten minutes. The contact time was over 10 minutes, and CO_2 adsorption capacity decreased. With the increasing adsorption time, the pores were filled with more CO_2 molecules, decreasing adsorption capacity. This circumstance can potentially approach a state of saturation, as discussed by Hauchhum and Mahanta (2014) and Xu et al. (2016). In addition, Xu et al. (2016) observed that the process of CO_2 adsorption by biochar is rapid within the initial 10 minutes. After that, the adsorption process occurred slowly and met the balance or saturation point in the 60th minute of adsorption. A research study must be undertaken to determine the saturation point in the adsorption time of biogas purification utilizing zeolite and sugarcane bagasse-based biochar. These studies reported that the C_2 adsorbed increased with prolonged adsorption time, continuing until the equilibrium or saturation point of the adsorbent was reached.

Fig. 2. *The impact of different contact times of adsorption on CO_2 level in biogas*

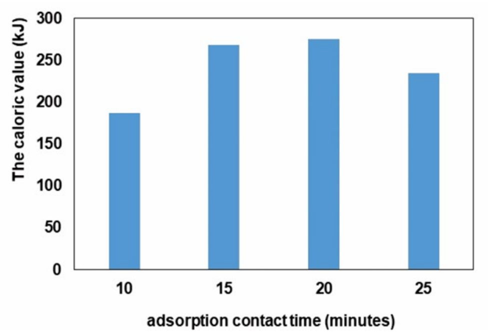


Based on observation, the blue flame shown by purified biogas was superior to that of non-purified biogas. The calorific value test of biogas in Fig. 3 showed significantly different results ($P < 0.05$). After CO_2 adsorption for 10 minutes, biogas had a calorific value of 186.83 ± 13.14 kJ. Meanwhile, after 15, 20, and 25 minutes, the calorific values were recorded as 267.63 ± 4.05 kJ, 275.72 ± 4.04 kJ, and 234.36 ± 13.26 kJ, respectively. The calorific value of biogas after 15, 20, and 25 minutes of purification showed a significant difference compared to the initial value after purification. Overall, there was a linear relationship between CO_2 adsorption content and the calorific value of biogas, except for biogas after purification for 25 minutes (see *Fig.3*). The result of the combustion efficiency is reported in *Table 5*, indicating that biogas subjected to CO_2 adsorption for 10, 15, 20, and 25 minutes achieved combustion efficiencies of 19.52, 31.66, 36.00, and 32.87% respectively. Notably, biogas performed the highest combustion efficiency after 20 minutes of CO_2 adsorption.

Table 5. *The decrease in CO_2 at different adsorption contact times*

Time (minutes)	The calorific value of biogas – Q (kJ)	Theoretical calorific value – LHV (kJ)	The combustion efficiency (%)
10	186.63 ± 13.14	956.64 ± 2.19	19.52
15	267.63 ± 4.05	845.34 ± 13.02	31.66
20	275.72 ± 4.04	789.89 ± 7.64	36.00
25	234.36 ± 13.26	713.30 ± 15.37	32.87

Fig.3. *The calorific value of biogas after CO_2 adsorption for 10, 15, 20, and 25 minutes*



The calorific value test checked the impact of biogas purification on the calorific value of biogas. The test was conducted by calculating the calorific value of 1 L of water boiled with biogas fuel. The biogas stove was ignited at the same level. Then, the increase in temperature and the decrease in the pressure gauge in the biogas tank were recorded every 30 seconds until the fire from the biogas stove went out. The calorific value of biogas indicates a greater amount of CO_2 being removed in biogas. After 20 minutes, the biogas exhibits its maximum calorific value. It occurred because after CO_2 adsorption on sugarcane bagasse-based biochar and zeolite for 25 minutes, the methane composition of biogas increased and impacted the calorific value.

We also tested the combustion efficiency of biogas. The combustion efficiency indicates the efficiency of biogas yield in generating heat energy. Biogas performed the highest combustion efficiency after CO_2 for 20 minutes. The results showed that the high calorific value of biogas is not directly proportional to high combustion efficiency. The primary factor affecting combustion efficiency is still the quantity of remaining gas in the biogas tank. The residual gas, which is non-combustible, cannot be adsorbed in biogas purification as complete combustion of biogas is not achievable. Another report also said that the difference in biogas's theoretical and empirical calorific value is due to the correction factor for energy loss associated with biogas stoves, which ranges from 15 to 25% (Pertiwiningrum et al., 2020).

Conclusions

This study found that the duration of adsorption contact had an impact on the reduction of CO_2 levels. Increasing the duration of adsorption led to a higher number of CO_2 adsorption points reaching saturation. The utilization of a combination of 50% sugarcane bagasse-based biochar and 50% zeolite (ZB2) in the process of CO_2 adsorption during biogas purification reaches its saturation point after 25 minutes of adsorption time. This study indicated that both the variations in the adsorbent's composition and the duration of contact influence the capacity of CO_2 adsorption. As the duration of adsorption contact increases, the capacity for CO_2 adsorption decreases. Further studies on the adsorption contact time should be replicated with a broader range of time variations exceeding 25 minutes to determine the saturation point of CO_2 adsorption by sugarcane bagasse-based biochar.

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