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CO_2 Methanation Over Empty Fruit Bunch (EFB)-Derived Biochar Catalysts: A Comparative Study with Nickel-Alumina

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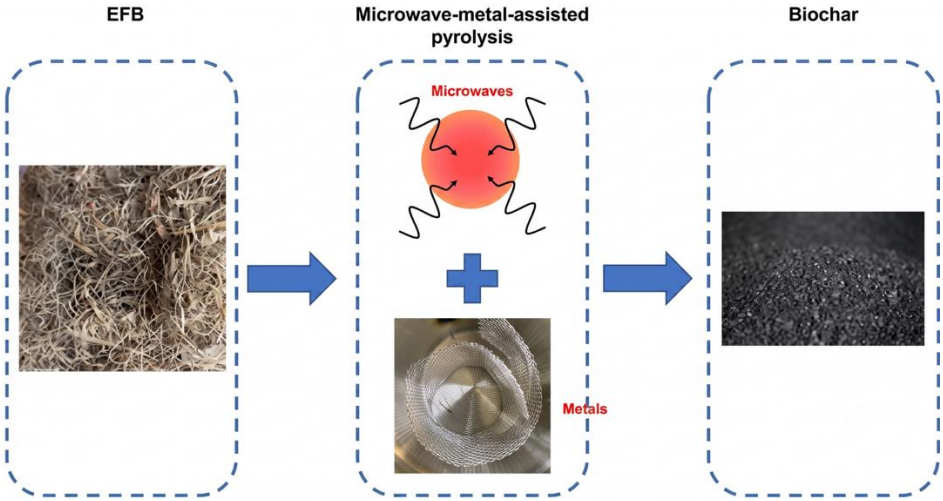
Graphical Abstract



The increasing demand for sustainable and cost-effective catalysts has driven research into alternative materials for CO_2 methanation. Conventional catalysts, such as nickel-alumina, are often expensive, have limited availability, and cause environmental hazards during production. This study aims to investigate the stability and durability of an oil palm empty fruit bunch (EFB)-derived biochar-based catalyst and compare its performance with nickel-alumina in CO_2 methanation. The methodology involved conducting Brunauer-Emmett-Teller (BET) surface area analysis on both catalysts, performing methanation reactions and analyzing the resulting gas composition. The expected outcome suggests that EFB-derived biochar demonstrates superior stability and conversion efficiency compared to nickel-alumina. This research highlights biochar's potential as a sustainable and economical catalyst alternative for CO_2 methanation, contributing to greener energy solutions.

Keywords: Biochar, methanation, methane production, catalyst, empty fruit bunch

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The methanation process of CO_2 is the process of creating methane (CH_4) from chemical compounds such as carbon dioxide CO_2 and hydrogen (H_2). This process has been getting attention for its potential to produce environmentally friendly energy and reduce the carbon footprint. The methanation process is important because it can reduce greenhouse gas emissions from the production, extraction, transportation, refining, and indirect emissions from the petroleum industry [1]. In carbon capture utilization and storage (CCUS), the methanation of CO_2 remains worth the manufacturing of methane, methanol, and renewable energy in the industry field by using catalyst conversion activity through various catalysts. Industry usually uses Nickel Alumina as catalyst generally expensive due to its availability and manufacture.

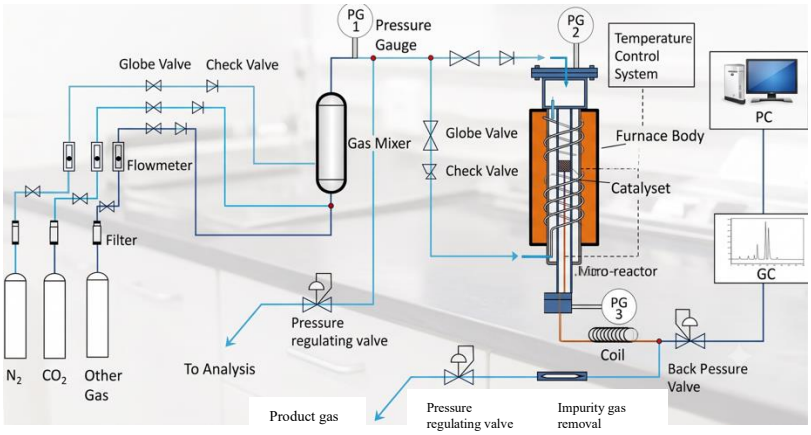

Figure 1. Low temperature methanation of fermentation gas with Nickel based catalyst in a multicomponent system [2].

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_20$$

The equation above is known as the CO_2 methanation reaction, which is also known as the Sabatier Reaction. Biochar as a catalyst support in CO_2 methanation is one of the possible techniques for increasing the productivity of methane during the process. The critical factors that define the catalytic performance are the conversion, the selectivity, and the stability of the catalysts. The Conversion rate determines how effectively CO_2 is converted to methane, while selectivity acts as a ratio of the amount of methane produced to other potential products. Stability relates to how the catalyst behaves and how long it can sustain its activity under the prevailing reaction conditions [3]. CO_2 activation is the most important step, where the molecule of CO_2 adsorbs to the surface of the catalyst, and a certain reduction process wherein the transfer of electrons either occurs from the catalyst metal to the CO_2 or in some instances, from CO_2 to the catalyst metal takes place. It involves breaking the C=O bond to increase the reactivity of the CO_2 in the hydrogenation process. After activation of CO_2, the hydrogenation steps are as follows – hydrogen addition steps where the hydrogen atom adds to the activated CO_2 molecule that results in the formation of methane (CH_4) and water (H_2O) [4].

EXPERIMENTAL

Material

Oil palm empty fruit bunch (EFB) was used as the precursor material for the biochar catalyst. The EFB was first dried to remove moisture, then weighed, with 30 g used for preparation, and subsequently pulverized into fine powder using a cutting mill machine. Nickel alumina catalyst was used as the comparison material and was already available in fine powder form; therefore, no additional preparation step was required before testing.

Method

Preparation of Empty Fruit Bunch (EFB) Derived Biochar Catalyst

The process began with drying the *Empty Fruit Bunch (EFB)* derived biochar to ensure the sample was in dry condition. The sample are weighted 30g before being sent to the cutting mill machine. The dried EFB-derived biochar was then pulverized into a fine powder using a cutting mill machine. Nickel Alumina catalyst did not require preparation since it was already in fine powder form.

Brunauer-Emmett-Teller (BET) Surface Area Analysis

Both catalysts began with weighting 6 grams before it was sent to the analysis lab to perform Brunauer-Emmett-Teller (BET) surface area analysis. This will provide details on specific surface area, pore volume and average pore size for both catalysts to determine their characteristic.

Methanation Process

Empty Fruit Bunch (EFB) derived biochar weighed of 6 grams and the catalyst was placed inside catalyst holder. The methanation system was injected by nitrogen gas N_2) for 10 minutes to ensure the system was free from unwanted gas. The valve tank was opened to inject 80 psi, 40 ml/min of H_2. The process continued by injecting 20 psi, 10 ml/min of CO_2. The catalyst holder monitored the pressure for 60 minutes. The valve was opened, and the effluent gas was collected into a gas collector bag. The gas collector bag was heated inside the oven at a temperature of 100°C for 60 minutes. The method was repeated using Nickel Alumina as catalyst.

Figure 2. Preparation catalysts, methanation process and gas analysis.

Gas Analyzer

The collected gas was sent to the gas lab to perform gas analysis. The gas analyzer will suck the gas collected to identify the gas components. The method was repeated using Nickel Alumina as a catalyst.

RESULTS AND DISCUSSION

This chapter presents the findings of the methanation process of the Empty Fruit Bunch (EFB) catalyst and Nickel Alumina catalyst, focusing on the performance of the CO_2 methanation and the stability and durability of the EFB biochar catalyst during prolonged reaction periods. Initially, Brunauer-Emmett-Teller (BET) Surface Area Analysis was conducted to measure the specific surface area. This test provides information about the specific surface area, pore volume and pore size distribution of the catalyst. By using the BET test, it helps to recognize the surface area available for catalytic reactions. Next, the methanation process was applied to get the product of methanation by injecting CO_2 and H_2 gas into the catalyst holder before collection inside the gas collector bag. Then, the gas is heated before being sent to lab analysis to perform gas analysis for our observation. The result is analyzed to determine CH_4 yield, CO_2 conversion rate and hydrocarbon presence.

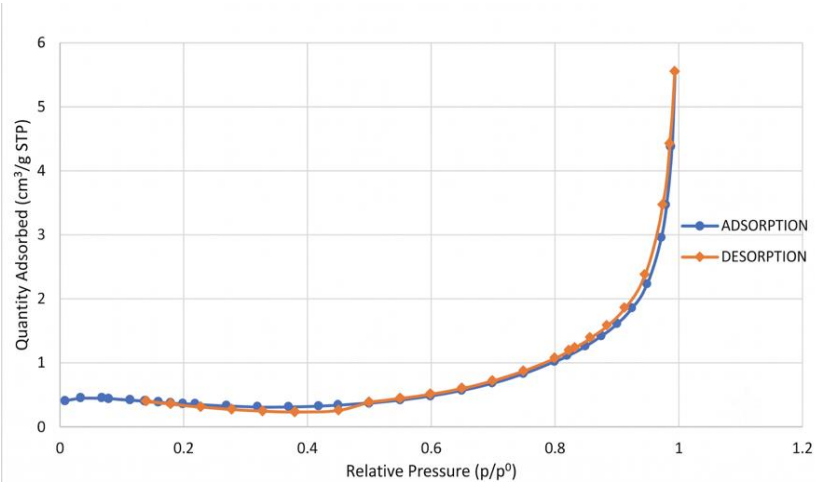

Figure 3. Graph isotherm linear plot EFB derived biochar catalyst.

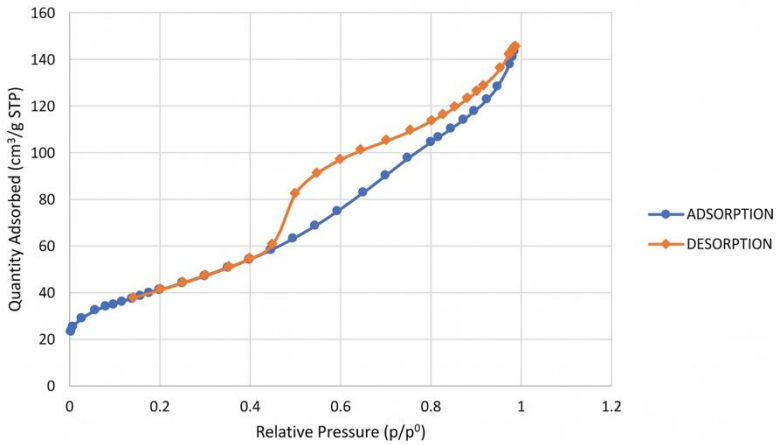

Figure 4. Graph isotherm linear plot Nickel Alumina catalyst.

Brunauer-Emmett-Teller (BET) Surface Area Analysis

For EFB-derived biochar, the microporous behavior can be seen at low relative pressures, which under (<0.3 p/po), indicates adsorption is very low and nearly flat. This behavior shows there is a small fraction of micropores in the structure of biochar catalysts. Next, the reading gradually increases in adsorption from 0.4 p/p⁰ to 0.8 p/p⁰. The mesoporous contribution makes the adsorption increase slowly. The sharp uptake at high 0.9 p/p⁰ and above shows pore condensation in macropores. This was influenced by typical mesoporous/ microporous materials, where gas molecules fill large pores at high relative pressures [5]. The desorption curve does not fully recollect the adsorption curve, forming a hysteresis loop. This indicates mesoporous structures with pore connectivity issues or ink-bottle pores. The graph shows Type IV or Type V isotherms, indicating a mix of micro and micropore and mesopores, with mesopores dominating the adsorption process [6].

For Nickel Alumina, the microporous behavior can be seen at low relative pressures, which under (<0.2 p/po) indicates adsorption is low. This behavior shows there was a small fraction of micropores in the structure of the catalyst (<2nm). Next, a sharp increase at high pressures of adsorption readings above 0.5 p/p° indicates the mesopores dominate the structure. The desorption curve does not follow the adsorption curve, confirming pore network effects. This indicates that some gas remains trapped in the pores. The gap between adsorption and desorption, called the hysteresis loop, shows a good catalytic application in methanation [7]. This indicates mesoporous structures. The high adsorption indicates that Nickel Alumina can hold a significant amount of reactant gases, which are CO_2 and H_2 to improve the efficiency. However, the large hysteresis loop shows some gas molecules remain trapped, which may slightly reduce desorption efficiency. The shape suggests a Type IV isotherm, which indicates the characteristic of mesopore material [6].

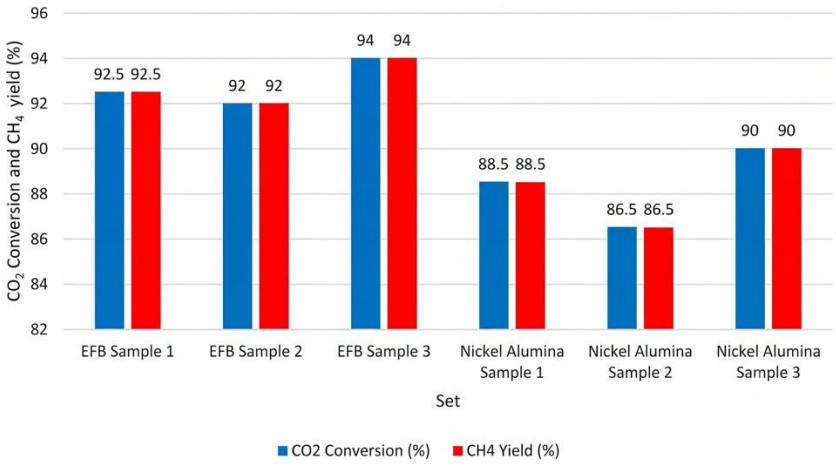

Figure 5. CO_2 conversion and CH4 yield.

Relation of CO_2 Conversion and CH_4 Yield

From the chart, it is evident that the EFB-derived biochar catalyst has higher CO_2 conversion of 92.5%, 92%, and 94% across samples 1,2 and 3. The CH_4 yield readings are 92.5%, 92%, and 94% for samples using the biochar catalyst. Sample 3 indicates the highest CO_2 conversion and CH4 yield at 94%, which indicates the highest conversion and yield observed, suggesting optimal catalytic activity under this condition. Meanwhile, sample 1 showed the lowest readings demonstrating the CO_2 conversion and CH_4 yield, indicating a slight drop in conversion and yield may indicate minor deactivation of active sites or reduced reaction rate. However, Nickel Alumina has lower CO_2 conversion of 88.5%, 86.5% and 90% for samples 1, 2 and 3. The CH4 reading are 88.5%, 86.5% and 90% showing lower than EFB-derived biochar catalyst. Sample 2 recorded the lowest reading of CO_2 conversion and CH4 yield. This set exhibits the lowest performance, possibly due to catalyst deactivation or reduced active sites. Sample 3 using Nickel Alumina, recorded the highest CO_2 conversion and CH4 at 90%.

The EFB-derived biochar catalyst performance makes it an ideal candidate for methanation compared to Nickel Alumina since it nearly equals value which is almost all the CO_2 are converted to CH4 which indicates a good selectivity using this catalyst. EFB-derived biochar performs well in methanation due to several factors. First and foremost, EFB-derived biochar catalysts have a large surface area and porous structure. This helps in improving the gas-solid in exchange and provides a more active site during this experiment (Hairy Azwan Bin Mohd Bakhtiar et al., 2019). Based on the result, it shows that the ratio of CO_2 conversion are almost 100% from all samples which shows a very low side product formed which is carbon monoxide (CO). Furthermore, EFB-derived biochar shows more stable performance since there is a minimal difference in CH4 yield across different samples, which indicates this catalyst is less affected by deactivation and maintains a stable catalytic function.

Compared to EFB-derived biochar, the performance of Nickel Alumina is more inconsistent due to it achieves low CO_2 conversion in all samples. The highest reading of CO_2 conversion and CH4 yield reading is 90% which is still lower compared to the lowest for EFB-derived biochar catalyst at 94% This shows that the Nickel Alumina catalyst is less effective in handling CO_2 methanation under the tested conditions. From the result, we can tell that the Nickel Alumina has lower CH4 selectivity since there is a decrease in reading in samples due to the presence of CO from the reading table. It indicates some CO_2 is converted into CO rather than CH4 due to incomplete CH4. In addition, there is the possibility of catalyst deactivation since Nickel Alumina is optimum used at higher temperatures from 300°C to 500oC [4]. Additionally, nickel-based catalysts are known to be susceptible to carbon deposition which leads to coking or blocking active sites over time that lead to reducing its efficiency [8].

Relation of Hydrocarbon and CO_2 Conversion

From the chart, it is evident that the CO_2 conversion is influencing the hydrocarbon reading (ppm) for each sample based on their types of catalyst. The relation between these results is important to study the effectiveness of different catalysts. The hydrocarbon reading indicates the presence of unreacted hydrocarbon or reaction gaseous types in that we get from the methanation process. The CO_2 levels represent the amount of CO_2 conversion. A lower CO_2 level should give a higher CO_2 conversion. Based on the graph, the hydrocarbon concentration at 2070 ppm, 1890 ppm is the lowest and 2150 ppm is the highest reading for samples 1, 2 and 3 using EFB- derived biochar as catalyst. Nevertheless, a lower reading of hydrocarbon shown when using Nickel Alumina corresponds across the samples with 1692 ppm, 1550 ppm as the lowest reading for this catalyst and 1800 ppm as the highest reading.

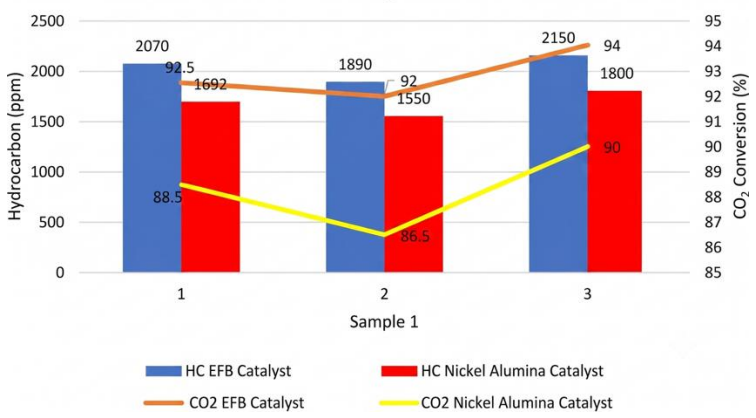

Figure 6. Hydrocarbon and CO_2 conversion efficiency comparison.

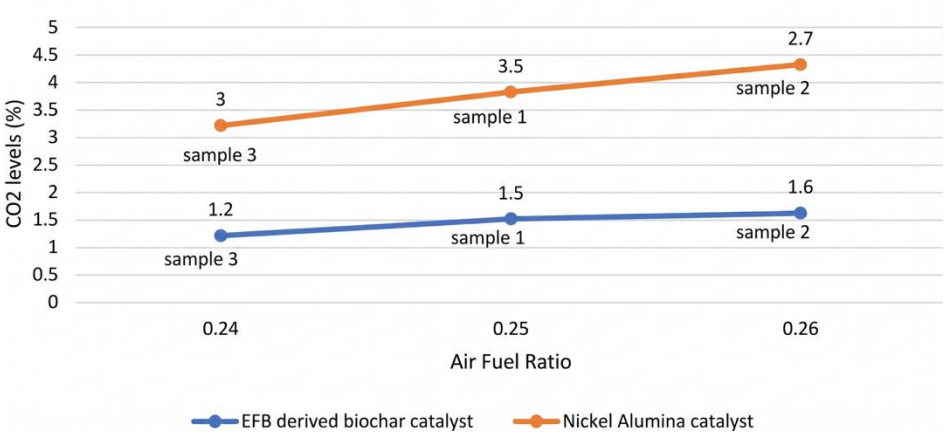

Figure 7. Relation of Air Fuel Ratio (AFR) with CO_2 concentration.

The influence of CO_2 levels is also significant to the CO_2 conversion for better catalytic activity understanding. According to the graph, using EFB-derived biochar, it shows that the CO_2 levels show the result of 92.5%, 92% and 94% for samples 1, 2 and 3. It shows that sample 3 shows the lowest CO_2 levels that indicate the best CO_2 conversion among all the samples. The observation of Nickel Alumina as a catalyst gives the result of CO_2 levels of 88.5%, 86.5% and 90% which indicate incomplete CO_2 conversion and lower catalytic activity compared to EFB-derived biochar. Sample 2 for Nickel Alumina set the highest value of CO_2 levels and sample 3 using the same catalyst gives the lowest value of CO_2 levels. EFB biochar consistently achieves lower CO_2 levels, particularly in sample 3 1.2%, suggesting superior CO_2 conversion efficiency 94%.

The result shows the EFB-derived biochar highlights lower CO_2 release with values of 1.2% (Sample 3), 1.5% (Sample 1), and 1.6% (Sample 2) across increasing AFR levels. The lower CO_2 release suggests that EFB biochar resulting higher CO_2 conversion efficiency which contributes to less CO_2 being retained in the system. The increase in CO_2 levels with AFR is minimal compared to Nickel Alumina, indicating that EFB biochar maintains consistent CO_2 reduction. This illustrates better catalytic performance and reaction optimization, ensuring CO_2 is efficiently converted into methane. On the other hand, CO_2 levels increase from 3.0% (Sample 3) to 2.3% (Sample 1) and 2.7% (Sample 2) as the AFR rises from 0.24 to 0.26 showing Nickel Alumina has higher CO_2 emissions. This is probably the cause of lower CO_2 adsorption efficiency that shows weaker catalytic activity, leading to an incomplete methanation process. The increase in CO_2 levels with AFR suggests that at higher oxygen availability. Therefore, the reaction influences towards CO_2 retention rather than conversion into methane.

Based on the result, EFB derives a biochar catalyst that is more effective because it has higher porosity and surface area. The larger porous structure provides better gas flow and more active site availability [9]. In addition, the chemical group on biochar provides better CO_2 capture which leads to higher conversion efficiency. Moreover, the minimal increase in CO_2 release suggests that EFB biochar remains catalytically active. Generally, Nickel Alumina as a catalyst shows higher CO_2 release compared to EFB-derived biochar because of its lower CO_2 interaction [10]. Nickel Alumina may have a Weaker interaction with CO_2, leading to incomplete conversion. During a prolonged period, Nickel Alumina may experience coking which reduces active sites and leads to less efficient CO_2 conversion that may cause possible catalyst deactivation [11]. Meanwhile, in the case of Nickel Alumina, the increase in AFR could be causing a shift in the equilibrium, favoring CO_2 retention over methanation.

CONCLUSION

In this study, the comparison of CO_2 methanation using EFB-derived biochar and Nickel Alumina catalyst has provided important knowledge into their efficiency, performance and best usage. This research uses different types of catalysts with the same condition for better comparison. The research addressed the primary objectives, where EFB-derived biochar exhibits promising characteristics as a catalyst for CO_2 methanation, highlighting its potential as a sustainable and effective alternative to conventional Nickel Alumina catalysts in carbon utilization technologies. In addition, EFB-derived biochar consistently shows better result in CO_2 conversion, CH_4 yield, hydrocarbon concentration, Air Fuel Ratio (AFR), CO_2 release compared to Nickel Alumina catalyst.

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