IJFAC

Indonesian Journal of
Fundamental and Applied Chemistry

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Synthesis Gas Production from Palm Oil Shell Waste using Al/Cr Metal Pillared Bentonite Catalyst for Eco-Friendly Fuel

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Abstract

The production of palm oil shell waste in the palm oil industry is increasing, but the utilization of palm oil shell waste has not been utilized optimally. Palm Oil Shells must be processed and utilized to reduce pollution and increase the use value of waste. This research aims to utilize palm oil shell waste using an Al/Cr metal pillared bentonite catalyst with a gasification process to produce synthesis gas. The bentonite pillarization process using Al/Cr metal aims to increase the catalyst surface area, basal distance and catalyst surface acidity so as to increase the catalytic activity of the catalyst in the gasification process. The gasification process is carried out at temperatures of 450, 500, 550 and 600°C. Adding the Al/Cr metal pillared bentonite catalyst, the H_2 concentration increased significantly. The effect of increasing temperature can increase the H_2 content by consuming CO within a certain time limit. Gasification process at temperature 600°C with a 20% Al/Cr metal pillared bentonite catalyst is the optimal condition for producing synthesis gas. The heating value of the gasification results in terms of the optimum HHV and LHV values is 16.21 MJ/Nm^3 and 14.59 MJ/Nm^3. Based on research, gasification of palm oil shells using an Al/Cr metal pillared bentonite catalyst is feasible for the production of environmentally friendly fuel.

Keywords: Palm Oil Shells, Al/Cr metal pillared bentonite, gasification, energy

Article Info

Received 22 August 2024
Received in revised 13 September 2024
Accepted 19 September 2024
Available Online 25 October 2024

Abstrak (Indonesian)

Produksi limbah cangkang kelapa sawit pada industri kelapa sawit semakin meningkat, namun pemanfaatan limbah cangkang kelapa sawit belum dimanfaatkan secara optimal. Cangkang Kelapa Sawit harus diolah dan dimanfaatkan untuk menurunkan pencemaran dan menambah nilai guna limbah. Penelitian ini bertujuan untuk memanfaatkan limbah cangkang kelapa sawit menggunakan katalis bentonit terpilar logam Al/Cr dengan proses gasifikasi untuk menghasilkan gas sintesa. Proses pilarisasi bentonit menggunakan logam Al/Cr bertujuan untuk menambah luas permukaan katalis, jarak basal dan keasamaan permukaan katalis sehingga dapat meningkatkan aktivitas katalitik katalis dalam proses gasifikasi. Proses gasifikasi dilakukan pada temperatur 450,500,550 dan 600°C. Penambahan katalis bentonit terpilar logam Al/Cr, konsentrasi H_2 meningkat secara signifikan. Pengaruh kenaikan temperature dapat meningkatkan kandungan H_2 dengan mengkonsumsi CO dalam batas waktu tertentu. Proses gasifikasi pada temperatur 600°C dengan katalis bentonit terpilar logam Al/Cr 20% merupakan kondisi optimal dalam menghasilkan gas sintesa. Nilai kalor pada hasil gasifikasi ditinjau dari nilai HHV dan LHV optimum yaitu 16,21 MJ/Nm^3 dan 14,59 MJ/Nm^3. Berdasarkan penelitian, gasifikasi cangkang kelapa sawit menggunakan katalis bentonit terpilar logam Al/Cr layak untuk produksi bahan bakar ramah lingkungan.

Kata Kunci: Cangkang Kelapa Sawit, bentonite terpilar logam Al/Cr, gasifikasi, energi

INTRODUCTION

Indonesia is currently experiencing an energy deficit due to its dependence on fossil fuels. Therefore, to overcome the problem of the energy crisis, it is necessary to diversify energy, namely looking for renewable alternative energy [1].

The production of palm shell waste in the palm oil industry is increasing, but its utilization is not yet optimal. Palm oil shells are agricultural waste that contain high levels of organic material and have the potential to be used as an alternative energy source. Palm oil shells have a higher volatile matter content of 72.49% compared to the volatile matter content in EFB of 54.13% [2]. Biomass with a high volatile matter content is suitable for conversion into synthesis gas and has the potential to produce high-gas products in the gasification process [3].

Gasification is a technology for processing solid waste into environmentally friendly gas (Syngas) consisting of H_2, CO and CH_4 gas [4]. Synthesis gas from conversion biomass through good gasification has high H_2 and CO content with low CO_2 content [5]. The reactions in the gasification process are as follows:
Water Gas Reaction:

$$C + H_2O \rightarrow H_2 + CO \quad (1)$$

Boudouard Reaction:

$$CO_2 + C \rightarrow 2CO \quad (2)$$

Shift Conversion:

$$CO + H_2O \rightarrow CO_2 + H_2 \quad (3)$$

Methanation:

$$C + 2H_2 \rightarrow CH_4 \quad (4)$$

The gasifier's operating temperature has an impact on how well biomass is converted throughout the gasification process. High-temperature efficiency will help with tar and carbon conversion, but it will cost a lot of money and consume a lot of energy [6]. In the meantime, it will result in issues with fuel conversion at low temperatures [7]. Therefore, a catalyst must be added to maintain high efficiency at low temperatures and reduce the amount of by products that are produced. The advantage of using an Al/Cr metal pillared bentonite catalyst is that it can increase the catalytic activity of the catalyst thereby producing optimal syngas in the gasification process.

MATERIALS AND METHODS

Materials

The main material in this research is palm oil shell combined with Al/Cr pillared bentonite catalyst and gasification medium (air and oxygen). Palm oil shell waste is taken from oil palm factory in Ogan Ilir, South Sumatra. The catalyst used is Al/Cr metal pillared bentonite catalyst.

Bentonite activation chemically and physically

Bentonite is chemically activated using H_2SO_4 then calcined in a furnace at a temperature of 400°C for 3 hours.

Al/Cr metal preparation

The Al/OH pillar solution was prepared by adding 80 mL of 0.5 M $AlCl_3$ to 240 mL of 0.5 M NaOH solution into a 500 mL beaker. Then the Cr/OH pillar solution was prepared by adding 80 mL of 0.5M $CrCl_3$ to 240 mL of 0.5M NaOH solution into a 500 mL beaker. The two solutions were mixed then stirred for 2 hours and left at room temperature for 48 hours.

Al/Cr metal pillarization of Bentonite

The combined Al/Cr solution was added to bentonite which had been activated chemically and physically, then stirred for 1 hour at room temperature, then filtered and washed with demineralized water. Al/Cr metal pillared bentonite was dried at 110°C for 4 hours.

Research Procedures

Palm oil shell waste is fed as much as 2 kg into the gasifier through the hopper without using a catalyst, then the valve is closed. Then the reactor is heated until the temperature reaches 450°C with the gasification temperature set at 450°C, 500°C, 550°C and 600°C. The gasification medium is injected into the bottom of the reactor and uses water to cool the condenser. This condensation process is carried out in the condenser using cooling water. The gasification process is carried out by injecting steam to supply the formation of H_2. After obtaining the gas product, the gasification gas is collected in a gas sampling bag for analysis. The gasification process is repeated using Al/Cr pillared bentonite catalyst.

Data Analysis

The gas results obtained from the gasification process are tested using a Gas Chromatography (GC) tool to obtain the composition of the gasification results. The Al/Cr metal pillared bentonite catalyst will be characterized using SEM, BET and XRD methods to determine changes in the structure of the catalyst.

RESULTS AND DISCUSSION

Scanning Electron Microscope (SEM) Analysis of Al/Cr Metal Pillared Bentonite Catalyst

SEM aims to analyze the morphology and surface structure of adsorbent particles and observe the pores or channels visible on the surface [8]. **Figure** 1 shows the results of SEM tests of Al/Cr metal pillared bentonite with various magnifications.

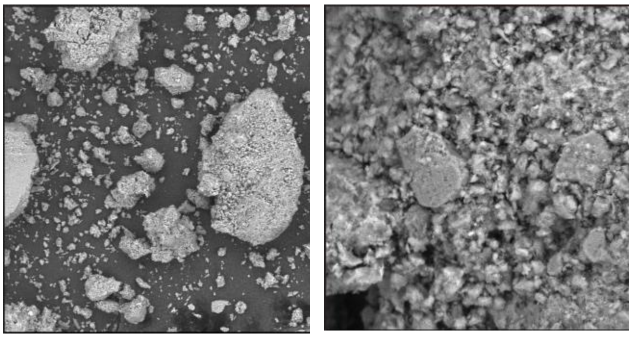

Figure 1. SEM Test Results for Pillared Bentonite Al/Cr Metal Magnification (a) 1000 times (b) 6000 times

The SEM results in **Figure** 1 show the surface morphology of Al/Cr metal pillared bentonite. Bentonite particles have a dominant size of 1µm. In the SEM test results, it can be seen that the pillarized bentonite has a rough structure. Thermal activation at 400°C can make the bentonite structure more porous. The SEM image of the pillared bentonite shows that the space between the layers has collapsed and produced a dense structure. The activation process in the form of pillarization affects the distribution of pores in Bentonite which allows the formation of new pores or changes to the existing micro and mesopore structure. The pore and mesopore structure influence the ability of bentonite to capture and store adsorbate molecules. Pore sizes that match the target molecule can increase the selectivity of bentonite.

Brunauer Emmet Teller (BET) Analysis Al/Cr Metal Pillared Bentonite Catalyst

BET analysis aims to identify the specific surface area, average pore size and pore volume of the sample. BET characterization was carried out on two samples, namely Bentonite before activation and Bentonite after Al/Cr metal pillaring as shown in **Table** 1.

Table 1. Characteristic Results with BET

Material	Area (m^2/g)	% Expansion
Bentonite Before Activation	45,35	120,26
Al/Cr Pillared Bentonite	99,89	

From the analysis results, the specific surface area of Bentonite after Al/Cr metal pillars has increased significantly. Bentonite has the ability to swell, where bentonite has interlayer spaces or has larger pores and a flexible structure so that pore engineering can be carried out to form pillars with micropore or mesopore sizes [9]. With this statement, the pillarization process can make the surface area of the Bentonite catalyst wider than the Bentonite catalyst before activation. The surface area of the catalyst will influence the activity of the catalyst, the greater the surface area of a catalyst, the more active phase will be distributed in the catalyst support, which will increase catalyst activity and increase product formation activity [10].

X-ray Diffraction (XRD) Analysis of Al/Cr Metal Pillared Bentonite Catalyst

XRD characteristics are used to study the structure and crystal size of solid materials. **Table** 2 shows the crystal size data from the XRD test results.

Table 2. Average Crystal Size Data

Peak 2θ	FWHM	Crytallite Size (Nanometer)
20,26	1,52	51,45
28,1	2,3	33,51
31,99	0,18	42,43
62,5	1,4	48,51
67,2	2,4	27,57
75,1	2,5	25,19
Average Size		38,11

Based on **Table** 2, it is found that the average size of Al/Cr pillared Bentonite crystals is 38.11 nm, where the crystal structure resulting from the pillarization method preparation process with Al/Cr metal has reached nano size so it can be concluded that the catalyst activation process carried out is going well. In the research of Onaola [11], the crystal size of bentonite before activation was 6.02 nm, so it can be concluded that there was a process of enlarging the crystal size which occurred due to the activation of bentonite.

Characteristics of Palm Oil Shells

Proximate and ultimate analysis was carried out to determine the characteristics of palm oil shell waste used as fuel. The volatile matter produced in palm oil shells is 72.49% [12]. The volatile matter content shows the ability of a material to evaporate. The volatile matter content of oil palm shells is greater than empty palm fruit bunches which have a volatile matter content of 54.13% [2]. High levels of volatile matter in biomass will speed up the heating process in gasification because volatile matter will heat the raw material in the combustion or oxidation zone [12].

The fixed carbon content in palm oil shells is 13.29% with a low ash content of 3.01%. Fixed carbon is closely related to volatile matter in determining the heating value of fuel. This is based on the balance that occurs between the amount of fixed carbon and volatile matter, where fixed carbon is the agent for producing heat and volatile matter is the combustion agent which can speed up the combustion process so that the resulting combustion time is shorter and the resulting calorific value is maximum. The calorific value produced is 22.11 MJ/Kg. High ash content can reduce the calorific value of biomass because it reduces the percentage of fixed carbon content so that it will have an impact on the calorific value of synthesis gas. The reduction in calorific value is because ash can inhibit the combustion process by reacting with oxygen so that the need for oxygen in the combustion process is reduced.

Effect of Temperature on Syngas Production Volume in Palm Oil Shell Gasification (Non-Catalytic)

The gas resulting from gasification from Palm Oil Shells is analyzed to determine the quality of the gas produced based on the composition of the gas volume. The gas analyzed consists of four main components, namely H_2, CO, CH_4 and CO_2. Based on the results shown in **Figure** 2, there is an increase in gas composition concentration along with the increase in gasification temperature (450-600°C).

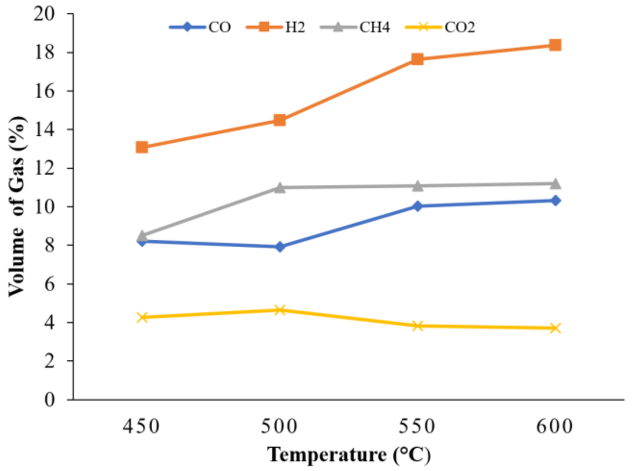

Figure 2. Effect of temperature on syngas production without a catalyst

In the gasification results, the highest concentrations of H_2 and CO were obtained at a temperature of 600°C of 18.37% (v/v) and 10.33% (v/v) Increasing the reaction temperature causes a hydrocarbon formation reaction which increases the H_2 concentration [13]. Higher temperatures support the production of increased H_2 concentrations due to the endothermic nature of the steam methane reforming reaction [14] and the conversion of volatile materials into gas occurs at high temperatures through endothermic reactions and steam cracking/reforming reactions [15]. From the gasification results, the CH_4 concentration obtained does not change significantly with increasing temperature, this is due to the limited methanation reaction [16]. **Figure** 2 shows the volume of syngas produced in the gasification process without using a catalyst.

The Effect of Al/Cr pillared bentonite on Syngas Production Volume of Palm Oil Shell Gasification

Figure 3 shows the volume of syngas produced in the gasification process using an Al/Cr metal pillared bentonite catalyst. In **Figure** 3, it can be seen that there is an increase in the H_2 and CO content along with increasing temperature. At a temperature of 600°C obtained the highest H_2 and CO composition of 40.89%(v/v) and 21.84%(v/v).

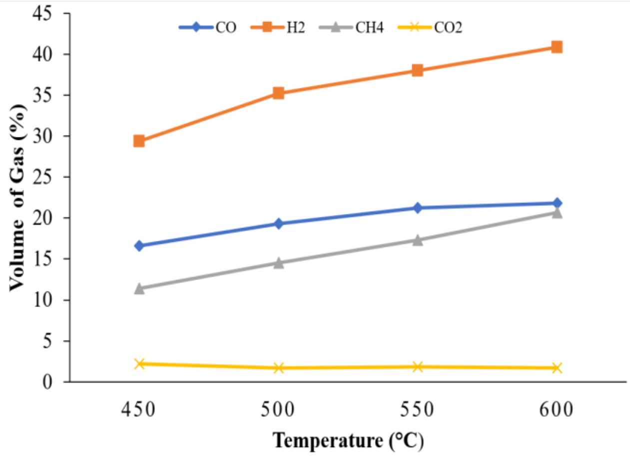

Figure 3. Effect of Al/Cr pillared bentonite on the volume of syngas produced in the gasification process.

The addition of Al/Cr metal pillared bentonite helps the gas decomposition process due to the catalytic cracking process. The catalytic cracking process will result in the formation of carbonium ions which help break down long hydrocarbon chains more easily [17]. In addition, the presence of Si and Al content in bentonite will increase the conversion reaction rate of complex compounds in Palm Oil Shells into low molecular gases. The SiO_2 and Al_2O_3 content in bentonite has an acidic side that can change the catalytic activity pathway to hydrocarbon cracking [18]. Where, the strong acid part of bentonite can increase deeper catalytic cracking [19]. By combining the two metals Al (Al_2O_3) and Cr (Cr_2O_3) in bentonite, basal spacing increases, resistance to high temperatures and increases the acidity of the catalyst, thereby increasing catalytic activity during the gasification process. The pillar metal bound to the interlayer affects the crystallinity of the bentonite, so that pillared bentonite has a more stable character and

DOI: 10.24845/ijfac.v9.i3.147

better heat resistance than bentonite without pillarization [20]. The addition of Al/Cr catalyst results in increased catalytic activity and affects the concentration of gas produced due to the catalytic cracking reaction, thereby increasing the production of syngas produced.

Palm Oil Shell Gasification Efficiency

Determination of gasification efficiency can be seen from the ratio H_2/CO, combustion gas ratio (CG/NCG) and carbon conversion efficiency (CCE). The H_2/CO ratio indicates the direction of use of the synthesis gas produced. The gasification efficiency of palm oil shells without using a catalyst or using an Al/Cr pillared bentonite catalyst can be seen in **Figure** 4-7. Based on research, the highest H_2/CO ratio without using a catalyst was found to be 1.83. However, when the temperature increases the H_2/CO ratio drops to 1.76. After Al/Cr pillared bentonite was applied to increase the main gas concentration yield, the H_2/CO ratio was in the range of 1.77-1.87. The use of bentonite provides a stable condition for the H_2/CO ratio. Synthesis gas with an H_2/CO ratio of around 1.0 is needed for the synthesis process in the production of aldehydes and alcohol, whereas an H_2/CO ratio close to 2.0 is needed for the production of fuel, ammonia and methanol. From the research results, the use of a catalyst produces an H_2/CO ratio close to 2.0 making it suitable for use as fuel. **Figure** 4 shows a comparison of the effect of temperature on the H_2/CO ratio of syngas from Palm Oil Shell Gasification without catalyst and using Al/Cr pillared bentonite catalyst.

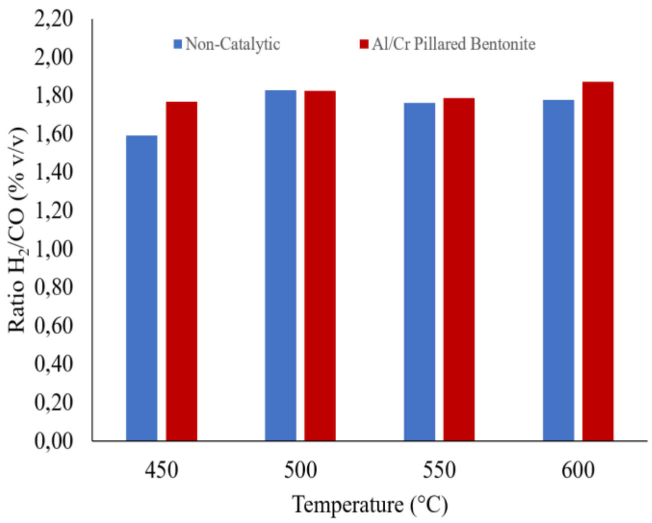

Figure 4. Effect of temperature on the H_2/CO ratio of syngas Gasification of Palm Oil Shells without a catalyst and the use of an Al/Cr pillared bentonite catalyst.

The CG/NCG ratio is an indicator of syngas quality. The ratio of combustible gas to non-combustible gas (CG/NCG) describes the ratio between combustible gas, namely H_2, CO and CH_4, to non-combustible gas such as CO_2, N_2 and O_2. The higher the ratio, the better the quality of the syngas produced. **Figure** 5 shows the highest CG/NCG ratio obtained from the gasification process using an Al/Cr metal pillared bentonite catalyst at a temperature of 600°C is 10.74. The CG/NCG ratio increases with increasing gasification temperature, this is proven by the increasing ratio at a temperature of 450°C to 600°C. **Figure** 5 shows the effect of temperature on the CG/NCG ratio of syngas from Palm Oil Shell Gasification without catalyst and using Al/Cr pillared bentonite catalyst.

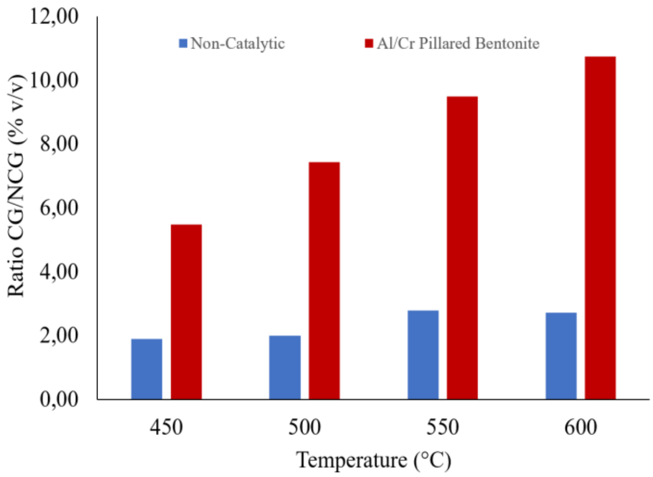

Figure 5. The effect of temperature on the CG/NCG ratio of syngas gasification of palm oil shells without a catalyst and the use of an Al/Cr pillared bentonite catalyst

The success of the gasification process is assessed by the performance and efficiency of the gasification. The parameters used to measure this are carbon conversion efficiency (CCE) and cold gas efficiency (CGE). **Figure** 6 shows the CCE in the gasification of Palm Oil Shells, where the highest CCE occurs at 600°C of 89.53% using Al/Cr metal pillared bentonite catalyst. From the research, it was found that the overall CCE increased along with increasing gasification temperature and catalyst use. Where increasing temperature increases the steam reforming reaction, which will encourage CCE. An increase in the temperature of the gasification reaction supports endothermic reactions such as the Boudouard reaction and the water-gas reaction which will consume the remaining carbon, triggering an increase in CCE [2].

Comparison of the effect of temperature on carbon conversion efficiency Gasification and cold gas efficiency of Palm Oil Shells without catalyst and using Al/Cr pillared bentonite catalyst can be shown in **Figure** 6.

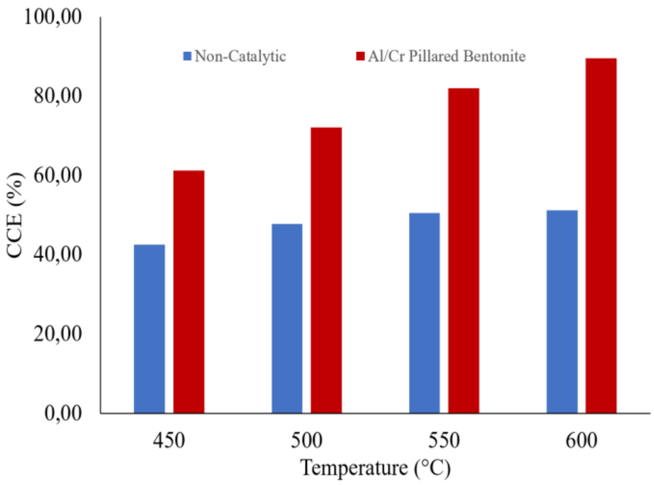

Figure 6. The effect of temperature on the carbon conversion efficiency of Palm Oil Shell Gasification without a catalyst and the use of an Al/Cr pillared bentonite catalyst

Cold gas efficiency (CGE) is the ratio of synthesis gas energy to raw material energy. The effect of temperature on cold gas efficiency of Palm Oil Shell Gasification without catalyst and using Al/Cr pillared bentonite catalyst is shown in **Figure** 7.

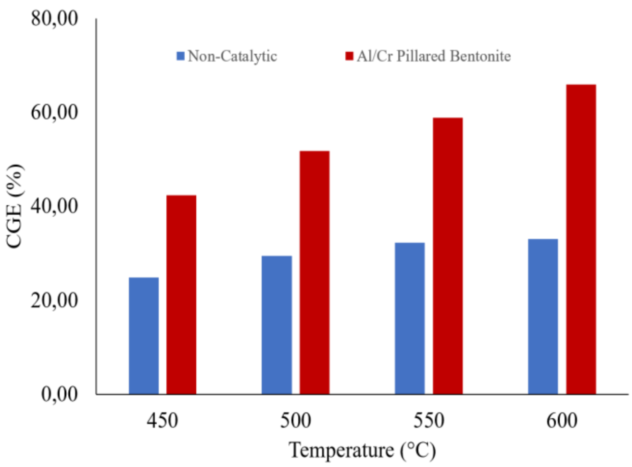

Figure 7. Effect of temperature on cold gas efficiency Palm Oil Shell Gasification without catalyst and using Al/Cr pillared bentonite catalyst

Cold gas efficiency (CGE) is the ratio of synthesis gas energy to raw material energy. In palm oil shell gasification, CGE increases with increasing temperature and catalyst use. The highest CGE is achieved at a temperature of 600°C was 65.97% using an Al/Cr metal pillared bentonite catalyst as shown in **Figure** 7. The increase in CGE can be attributed to a decrease in the amount of CO_2 and an increase in the amount of CO and H_2 from the gasification results. According to research of Su [21], the content of volatile substances and fixed carbon can also influence the % CGE produced.

Calorific Value of Syngas from gasification of palm oil shells

Calorific value is the energy released when biomass is completely burned. The heating value of syngas is calculated using the HHV and LHV equations [22]. The HHV and LHV values resulting from gasification are shown in **Figures** 8 and 9. From the research results, it was found that the maximum heating value in the gasification of palm oil shells was obtained by the highest HHV value at a temperature of 600°C uses Al/Cr metal pillared bentonite of 16.21 MJ/Nm^3. An increase in temperature causes the heating value to increase due to the promotion of tar cracking and hydrocarbon reformation reactions. Increasing the reactivity of the water-gas reaction with an increase in reaction temperature can accelerate the production of H_2 and CO and increase the heating value of syngas. HHV of syngas at high gasification temperatures is influenced by the increase in CO and H_2 moles [23]. The effect of temperature on the HHV of syngas from gasification of Palm Oil Shells without catalyst and the use of Al/Cr pillared bentonite catalyst is shown in **Figure** 8.

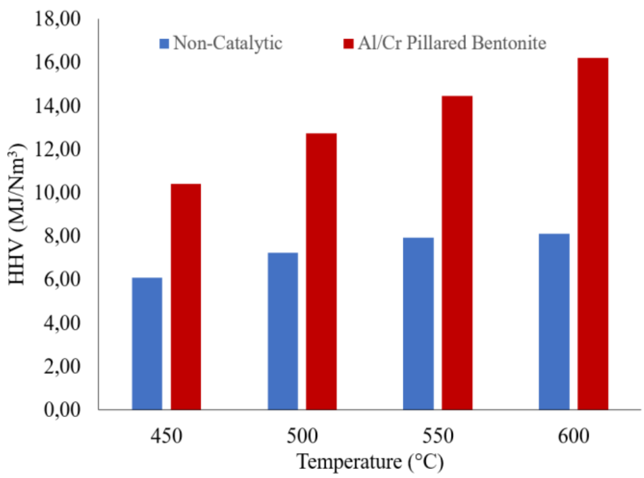

Figure 8. The effect of temperature on the HHV of syngas resulting from the gasification of Palm Oil Shells without a catalyst and the use of an Al/Cr pillared bentonite catalyst

LHV of syngas is calculated from the concentration of CO, CH_4 and H_2 of syngas. **Figure** 9 shows a comparison of the effect of temperature on the LHV of syngas from gasification of Palm Oil Shells without catalyst and the use of Al/Cr pillared bentonite catalyst. In **Figure** 9, the highest LHV value is at a temperature of 600°C using Al/Cr metal pillared bentonite of 14.59 MJ/Nm^3.

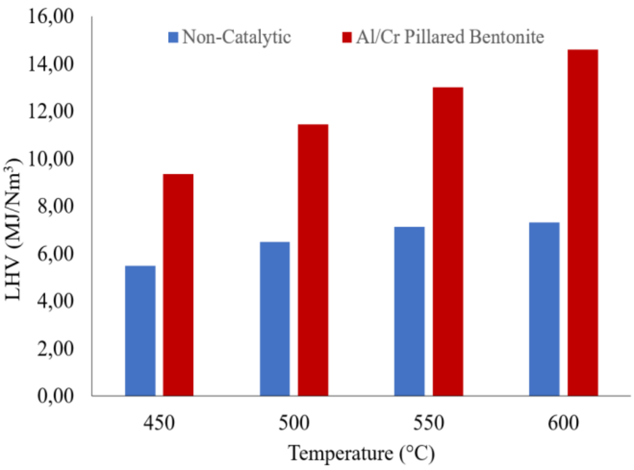

Figure 9. The effect of temperature on the LHV of syngas resulting from the gasification of Palm Oil Shells without a catalyst and the use of an Al/Cr pillared bentonite catalyst

CONCLUSION

The use of a catalyst has a significant influence on the syngas yield produced. The volume concentration of syngas increases as the amount of catalyst increases. From the research results, the largest concentration of H_2 and CO values was achieved using Al/Cr pillared bentonite at a temperature of 600°C was 40.89 (v/v) and 21.84 % (v/v), H_2/CO ratio was 1.87, CG/NCG ratio was 10.74, CCE was 89.53% and CGE was 65.97%. The heating value produced by HHV and LHV is 16.21 MJ/Nm^3 and 14.59 MJ/Nm^3. The addition of an Al/Cr bentonite catalyst can increase the heating rate of biomass, this is because SiO_2 and Al_2O_3 in bentonite have high thermal conductivity and the pillarization of Al/Cr metal in bentonite results in increased basal spacing and increases the surface area of the catalyst from 45.35 m^2/g becomes 99.89 m^2/g, there by accelerating the catalytic cracking reaction and optimizing biomass decomposition, there by producing more syngas.

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