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Key words: *bentonite, fine coal, fixed bed gasifier, producer gas*
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SYNGAS DERIVED FROM CATALYTIC GASIFICATION OF FINE COAL WASTE USING INDONESIAN POTENTIAL CATALYST

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Fine coal waste from the coal mining process has not been used as clean energy even though the amount is very abundant in the world. The conversion of fine coal to syngas is a new way to increase the value of fine coal. Syngas composition, gas ratio, gasification efficiency, and heating value of syngas have been determined under various conditions of temperature (550-750 °C) and bentonite catalyst ratio (0-0.25). The results indicate that fine coal is the suitable raw material for the gasification process. The increase in temperature has increased the volume percentage of H_2. At the highest temperature (750 °C), the gas composition consists of 42.6 vol% H_2, 19.1vol% CO, 19.5 vol% CH_4, and 7.9vol% CO_2. The best performance was achieved when the catalyst/feed ratio is 0.25 with the gas composition of 54.3vol% H_2, 26.2vol% CO, 23.8 vol% CH_4, and 3.5vol% CO_2, heating value and gasification efficiency were 19.72 MJ/Nm^3 and 72.27% at 750 °C.

Key words: bentonite, fine coal, fixed bed gasifier, producer gas

INTRODUCTION

The increase in world population causes the world's energy demand to increase as well. Currently, 96% of hydrogen and electricity needs are still dominated by fossil energy [1]. Of the several types of energy available, coal is an abundant and cheap fuel. The use of coal accounts for a third of total energy utilization globally, particularly on electricity generation (40%). Based on energy availability, the world's stored coal reserves can supply for 153 years, much longer than petroleum (50.6 years) and natural gas (52.5 years) [2].

Coal mining with a continuous open pit system has produced several by-products, including fine coal. Fine coal measuring <3 mm, which is formed due to the crushing by the unit working on the coal seam during mining operations [3]. Refined coal after cleaning is important in restoring an economically valuable energy source and preventing environmental pollution [4]. Fine coal is produced between 5-10% of the total coal production as a whole. Particulate fine coal, when inhaled, has the potential to cause respiratory diseases and other health problems. Fine coal contains high sulfur, so its use through direct combustion will be harmful to the environment [5]. The coal processing industry (production, transportation, and handling of lumped coal produces millions of fine coal particles during mining operations and is often regarded as waste [6]. This waste has enormous potential to be used as an energy raw material. The utilization of fine coal will not only add value to it but also serves to reduce environmental pollution.

The use of coal has resulted in high carbon dioxide emissions. The world's attention to the worsening environmental condition due to the emission of harmful gases such as CO_2, SOx, and so on encourages clean coal technology. In the mining industry, fine coal is used as fuel for boilers and power plants to meet the energy and electricity needs of a mining company [7]. Several studies on fine coal are known to convert fine coal into briquettes. Balraj et al. [8] and Manyuchi et al. [9] used fine coal and waste biomass to make briquettes at various mixtures and adhesives ratios.

As far as the research team has studied, the use of fine coal to produce syngas has never been done. Fine coal gasification is a technology that can be applied to increase the use-value of fine coal as a cleaner fuel. Therefore, fine coal is very interesting to study in the conversion process into environmentally friendly syngas. The purpose of this study is to evaluate the syngas produced from the fine coal catalytic gasification process.

MATERIALS AND METHODS

Materials handling

Fine coal is used as materials to feed the gasifier. Fine coal was obtained from open-pit coal mining in Tanjung Enim, South Sumatera, Indonesia. Fine coal was a waste from the coal mining process that has been cleaning. After cleaned and sieved to 0.5 mm, fine coal is ready to be fed into the gasifier. The proximate and ultimate analysis was performed to determine the physical and chemical properties of fine coal.

Bentonite is used as the catalyst for the gasification process. Bentonite was obtained from Sarolangun village, Jambi Province, Indonesia. Bentonite was activated us-

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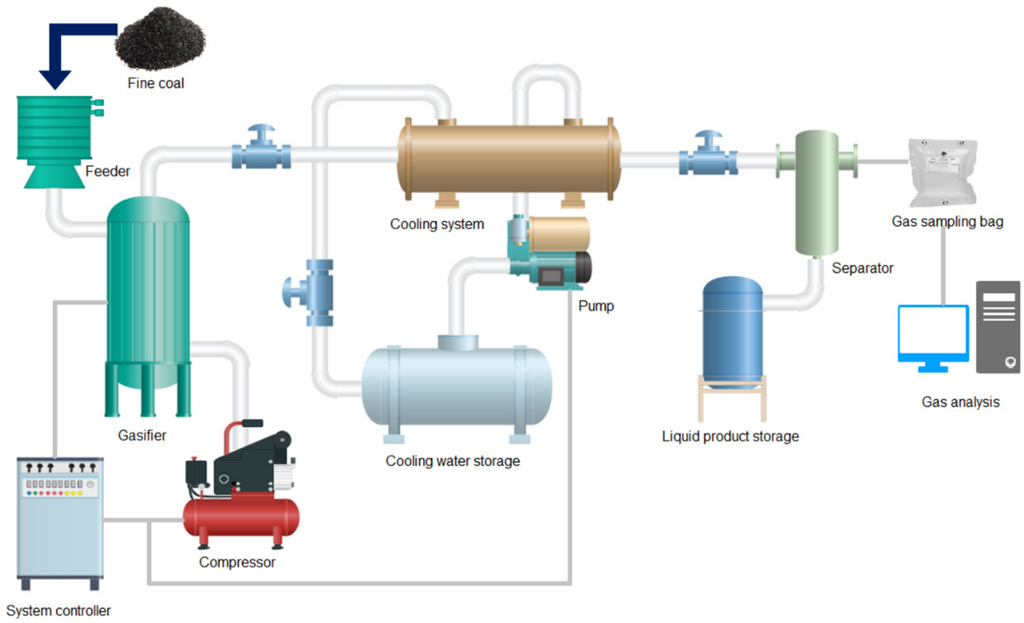

Figure 1: Flow Diagram of Fine Coal Catalytic Gasification

ing thermal activation method by muffle furnace (Thermo scientific thermolyne F48050-33). The chemical composition of natural bentonite was determined using XRF.

Experiments

The fine coal gasification was performed in a modified fixed bed reactor. The gasifier apparatus has been illustrated in Fig. 1 according to the previous work [10]. The equipment consisted of the gasifier with an electrical heater, cooling system, pump, valve, feeding system, sampling port, and control system. For safety, the body of the reactor was insulated by glass wool. The gasifier was made of stainless steel (SS316) with diameter and length of 0.23 m and 0,67 m. The nozzle heater was placed on the reactor wall. Two K-type thermocouples were positioned at the top and bottom of the reactor and connected to the control panel. Before the experiment was begun, fine coal was placed inside the feeding system. To start, the gasifier was set to reach the specified temperature as the independent variable. The temperature was set to 550, 650, and 750 °C of each gasification process in this study. The process took place approximately 30-60 min and was considered as residence time. After the process reached a stable and steady-state condition and the gas temperature has been lowered through the cooling system, sampling was carried out. The producer gas was gathered in a gas sampler bag. The composition of gas was then further analyzed. To evaluate the catalytic effect on fine coal gasification, Bentonite is applied to the gasifier together with fine coal at high temperature (750 °C) with a variation of the catalyst to feed (C/F) ratio of 0, 0.125, and 0.25 and analyzed.

Analysis of producer gas

The gas product was analyzed using Perkin Elmer Clarus 680 Gas Chromatography equipped flame ionization detector (FID). He and N_2 were used as the carrier gas. In order to know the quality of the gas product, the heating value of gas was calculated using equation 1-2. According to Tian et al. [11], gasification efficiency is analyzed based on efficiency of cold gas and carbon conversion. Furthermore, the ratio between hydrogen and carbon monoxide and the ratio between combustible and non-combustible gas were evaluated. The heating value (HHV and LHV) of producer gas was determined using the following equation (Eq. 1 and 2) [12]:

$$LHV=\left(\left(25.7\times H_2\right)\left(30\times CO\right)+\left(85.4\times CH_4\right)\right)\times 0.0042 \quad (1)$$

$$HHV=\left(\left(30.52\times H_2\right)\left(30.18\times CO\right)+\left(95\times CH_4\right)\right)\times 0.0041868 \quad (2)$$

HHV and LHV are lower and high heating value (MJ/Nm^3); H_2, CO, and CH_4 are percentages of hydrogen, oxidant carbon, and methane (vol%), respectively.

RESULTS AND DISCUSSION

Materials characteristics

Fine coal is coal waste that is mostly used as fuel for steam power plants. Fine coal characteristics have been identified through proximate and ultimate analysis, which refers to the ASTM standard method (Table 1). To facilitate the characteristic analysis, fine coal is compared with raw coal.

Based on the analysis, fine coal as raw research material is included in the 5800 calorie coal criteria. The sulfur content in fine coal was <0.5%, indicating that fine coal is suitable for the gasification process [14]. The carbon content of fine coal was above 60% and considered high [15]. This percentage was not too far from the content of the lump coal. The same thing happened to the volatile matter content, which was lower than raw coal [16].

The percentage of fixed carbon fine coal was lower than raw coal because fine coal was a waste from the coal

Table 1: Fine Coal Characteristics based on Proximate and Ultimate Analysis

Parameter	Standard Method	Fine coal	Raw coal [13]
Total moisture (%)	ASTM D 3302/3302M-17	13.48	3.94
Ash content (%)	ASTM D 3174-12	4.10	1.87
Volatile matter (%)	ASTM D3175-17	41.18	23.26
Fixed carbon (%)	ASTM D 3172-13	41.25	70.93
Total Sulphur (%)	ASTM D 4239-2016	0.42	0.20
HHV (MJ/kg)	ASTM D 5865-2013	28.18	30.64
Carbon (%)	ASTM D5373	64.05	80.66
Hydrogen (%)	ASTM D5373	4.43	4.18
Nitrogen (%)	ASTM D5373	0.90	0.85
Oxygen (%)	ASTM D5373	12.63	20.97

mining process, but it was much higher than biomass such as corn cob, coconut shell, palm kernel shell, palm empty fruit bunch, and others. [17]–[19]. Fixed carbon in a material affects the HHV value of the fuel [20]. Fixed carbon is 40–80% in the range in raw coal, depending on its quality [21]. The ash content of fine coal was <5%, which indicates that fine coal is suitable for the gasification process. High ash can reduce HHV because ash is a non-combustible material. The inorganic energy requirements of forming ash for thermal breakdown and the transition phase are taken from the combustion energy of fine coal and cause a reduction in the caloric value [22]. In contrast to raw coal, the ash content of fine coal is higher [23], [24]. The ultimate fine coal analysis results stated that the carbon value reaches 64.05% (as received). Carbon was closely related to heating value. Carbon and hydrogen in fine coal would later contribute to the formation of CO and H_2, which are the main components of synthesis gas.

Bentonite was acted as the catalyst in the gasification process obtained from Sarolangun, Indonesia. The compositional analysis was carried out to find the chemical compositions of bentonite by X-ray fluorescence (XRF) (Table 2). Bentonite consisted of a large number of SiO_2 dan Al_2O_3. The presence of SiO_2, Al_2O_3, Fe_2O_3, and MgO as the dominant components showed their potential to catalyze the gasification process. Based on Qin et al. [25], alkali metals have been shown to influence the gasification process.

Table 2: Chemical composition of natural bentonite by XRF

Chemical composition	Value (%)
SiO_2	71.12
Al_2O_3	20.77
Fe_2O_3	3.13
MgO	2.12
CaO	1.30
P_2O_5	0.85
K_2O	0.32
TiO_2	0.13

Effect of temperature on producer gas composition and ratio

Temperature is one of the significant parameters in the gasification process. In this study, fine coal gasification was performed at distinct temperatures of 550, 650, and 750 °C. The influence of temperature on the composition and ratio of gases has been studied. An increase in temperature impacts the rise in H_2 but decreases the volume fraction of other gases (CO, CH_4, CO_2), as shown in Fig. 2. The concentration of H_2 reaches 42.6 vol%. Hydrogen increased by 12.4 vol% since the temperature rose to 750 ºC, which was previously only 30.2 vol% at 550 ºC. The CO concentration has initially been 23% at 550 °C but then decreased with the rise in gasification temperature to 19.1 vol%. Initially, carbon from fine coal reacted with oxygen both from the gasification agent and fine coal (partial oxidation). Then, the partially formed CO reacts with the steam from the pre-drying phase to form CO_2 and H_2.

Methane is a combustible gas found in gasified gas but is not included in the syngas category. At 550 °C, the highest content of CH_4 was achieved at 23.7 vol% by methane formation reaction in the reduction zone. Methane re-reacts with carbon dioxide to form H_2. As the temperature has risen to 750 °C, the CH_4 concentration fell as well as CO_2, which resulted in a significant increase in the H_2 concentration. The reaction is known as dry reforming methane in the reduction zone. The CO_2 content remained in the final gas composition at the highest temperature but has decreased by 4.5 vol% from the initial concentration of 12.4 vol% to 7.9 vol%.

This trend occurs because CO_2 production came from shifting and cracking reactions, which are influenced by water-gas shift reactions and tar cracking. At the same time, the Boudouard reaction consumed most of the CO_2 that has been generated. Furthermore, because the CO_2 production rate is lower than the consumption rate, the CO content decreases with increased gasification temperature. The hydrogen concentration slightly increased with the rise of gasification temperature influenced by the increase in the reaction of water-gas and tar breakdown. The increas-

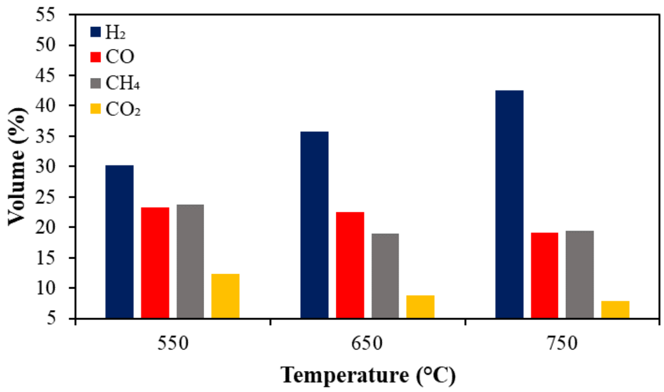

Figure 2: Temperature-influenced Syngas Composition

ing gasification temperature encourages the Boudouard and water-gas reactions to increase the CO even more. Hereinafter, rising the temperature in the gasification process also encouraged CH_4 (vapor phase) reaction and reduced CH_4 concentration. Le Chatelier's principle can also explain this trend that the endothermic reaction is suitable for product formation at higher temperatures. Syngas is produced mainly from the char partial oxidation influenced by water-gas and Boudouard reactions.

The H_2/CO ratio of the gas described the use of the gas produced. H_2 and CO concentrations were obtained through the analysis of gas composition in the previous section. Fig. 3 shows the effect of temperature on the H_2/CO and CG/NCG ratio. Both gas ratios have risen along with the increase in gasification temperature. Since the rise on hydrogen was greater than that of carbon monoxide, the ratio of H_2/CO increased by degrees. If the gasification temperature was relatively low, CO_2 tended to be generated through water-gas shift reaction and methanation reaction (exothermic) [26]. For fine coal gasification in this study, the H_2/CO ratio was 1.3-2.23. The production of chemicals such as methanol uses the H_2/CO ratio of 1-2 that fitted this study results.

Fine coal has shown its potential as a fuel to produce syngas with a higher ratio of H_2/CO at 750 °C. This was related to the small diameter of fine coal to have a smoother flow in the reactor. The greater the surface area of the material (fine coal), the more conductive heat transfer increases according to Fourier's Law. According to Madadian et al. [27], the carbon conversion rate can be increased if the conductivity is large due to the larger surface area of the feedstock. The CG/NCG ratio stated the ratio between combustible gas and non-combustible gas. Combustible gas in producer gas consists of CO, H_2, and CH_4, while non-combustible gas consists of CO_2, O_2, and N_2, resulting from gasification agents. As illustrated by Fig. 2, the CG/NCG ratio increased with increasing temperature. A significant result obtained when the temperature raised from 650 °C to 750 °C. The highest CG/NCG ratio achieved was 4.83. Furthermore, higher temperatures have also been able to reduce CO_2 gas so that the ratio is high. This CO_2 reduction occurred according to the dry reforming reaction of methane, evidenced by the decrease in CH_4 concentration when the temperature increases.

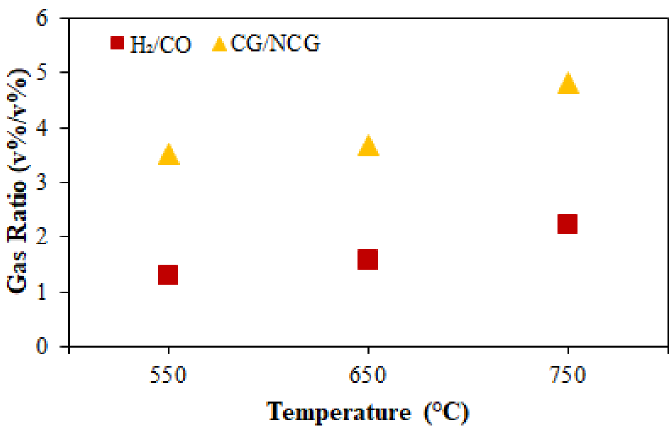

Figure 3: Effect of Temperature on H_2/CO and CG/NCG Ratio

Effect of temperature on gasification efficiency and heating value

The effect of gasification temperature on the heating value of syngas illustrated in Fig. 4. The temperature change shows a slight decrease in HHV and LHV of syngas. The HHV and LHV at 550 °C are 16.23 MJ/Nm^3 and 14.70 MJ/Nm^3. These results are associated with changes in the concentrations of CH_4, H_2, and CO, which are not significant to the temperature where the methane concentration decreases with increasing reaction temperature. These results also confirm that the produced gas can be used as fuel gas for the Fischer-Tropsch synthesis.

LHV and HHV decreased in consequence of the decrease in carbon monoxide and methane concentration from the producer gas. The concentrations of CH_4 and CO have a more generous contribution to the heating value of gas. The system showed good performance in terms of heating value, which can be seen from the slight heat reduction. Cold gas efficiency can be recognized as the ratio of the energy produced by the producer of gas to the calorific value of fine coal. The efficiency of cold gas determined gasification performance. It relied on the LHV of syngas compared to fine coal, which is constant during the gasification process. Gasifier design, raw material,

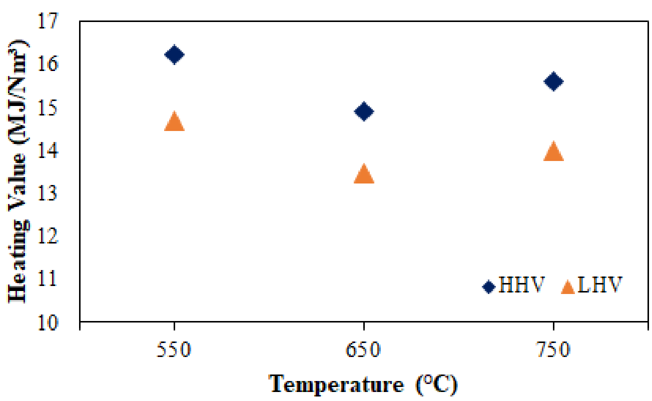

Figure 4: Effect of Temperature on the Heating Value

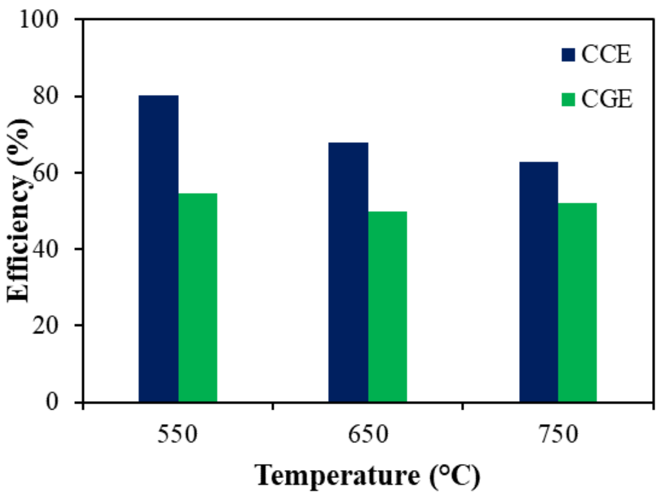

Figure 5: Effect of Temperature on Gasification Efficiency

moisture content are parameters that affect CGE. Fig. 5 illustrates the influence of reaction temperature on CCE and CGE. As observed, CCE and CGE had declined when the temperature raised from 550 to 650 °C, but then increased slightly when the temperature rose to 750 °C because the syngas produced had a higher H_2 content compared to CO and CH_4. At the end of the process, CCE and CGE were 62.81% and 51.90% at 750 °C.

Effect of catalyst on syngas composition

From the previous section, the most influential temperature which made hydrogen continue to increase occurred at 750 °C. Therefore, investigations of the catalyst performance on the syngas were continued at 750 °C. The hydrogen concentration increases as the catalyst to feed ratio (C/F) increases. The highest hydrogen concentration was obtained in increments of 0.25 C/F (54.3% vol). Fig. 6 demonstrates that the leading combustible gases (H_2, CO, and CH_4) were increased by rising the C/F. The CO_2 content was reduced with the higher addition of bentonite.

In the catalytic gasification process of fine coal, the volume percentage of main gasses in gas increased after bentonite is applied. Initially, at the highest temperature, hydrogen was only able to produce 42.6%, but after 0.125 C/F and 0.25 C/F was applied, H_2 concentrations increased to 46.6% and 54.3%. H_2 increased by 11.7 vol% at the same temperature after the bentonite was added. Furthermore, the CO_2 content was successfully reduced from 7.9% to 3.5%. The synergy effect between increasing gasification temperature and bentonite has a positive impact on increasing gas concentration. Bentonite consists mostly of SiO_2 and Al_2O_3. According to Oliveira et al. [28], both compounds have good heat conductivity. Besides, Fe_2O_3 in bentonite can increase the char gasification process [29]. Furthermore, some literature has reported that K_2O, Na_2O, CaO, MgO, and Fe_2O_3 in material play an important role in the catalytic effect during the gasification process. Fe_2O_3 and CaO were active components that have a catalytic impact on the gasification process.

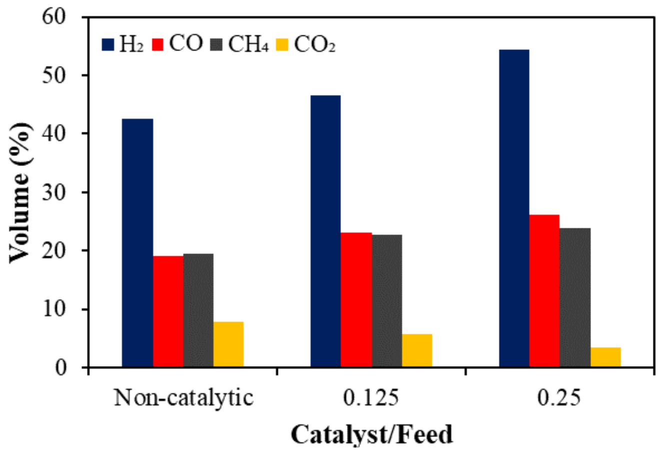

Figure 6: Effect of catalyst on producer gas composition

The H_2/CO ratio described the use of the syngas produced. The different properties of syngas are required for further chemical production. For example, syngas with an H_2/CO ratio of approximately one is necessary to synthesize oxo in aldehyde and alcohol production. The ratio of H_2/CO approach to 2 is required for fuel production, Fischer-Tropsch synthesis, and methanol [30]. In fine coal gasification, both original and catalytic, it has been carried out to determine the H_2/CO ratio (Fig. 6). The highest H_2/CO ratio was obtained in the gasification process without using a catalyst (2.23). After bentonite was applied to increase the main gas concentration, the H_2/CO ratio decreased but remained in number 2. Bentonite provided a stable state to the H_2/CO ratio. Syngas produced through a catalytic process using bentonite is suitable for use as fuel.

The CG/NCG ratio provides an overview of the comparison between combustible gas (CO, H_2, and CH_4) and non-combustible gas (O_2, N_2, and CO_2). It can be seen that by adding a catalyst, the CG/NCG ratio increases significantly (Fig. 7). This is consistent with the composition of the gas that has been analyzed, where the CO_2 concentration continues to decrease with the addition of bentonite. The highest CG/NCG ratio was obtained at 0.25 C/F of 11.46.

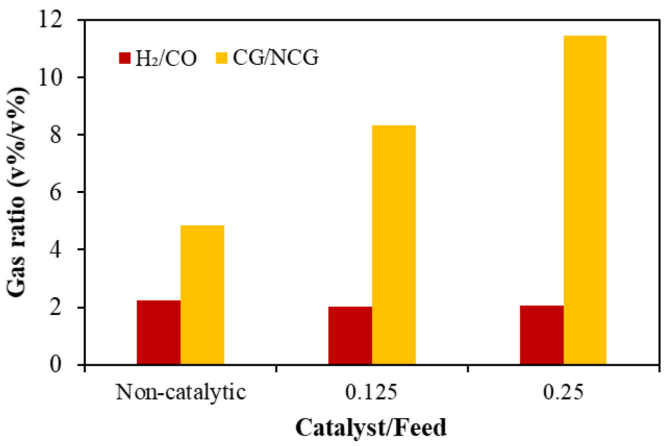

Figure 7: Effect of Catalyst on H_2/CO and CG/NCG Ratio

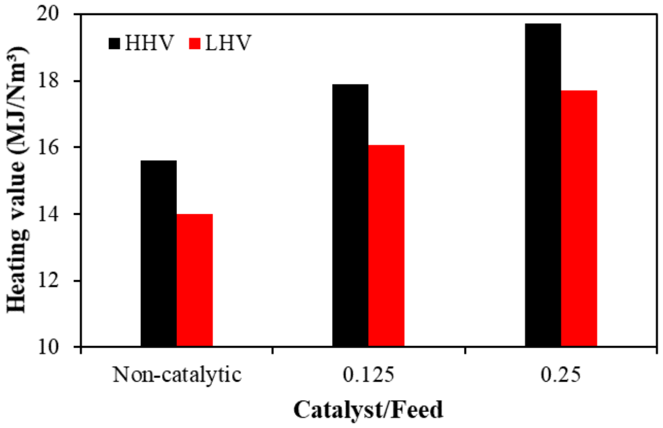

Figure 8: Effect of Catalyst on HHV and LHV of Syngas

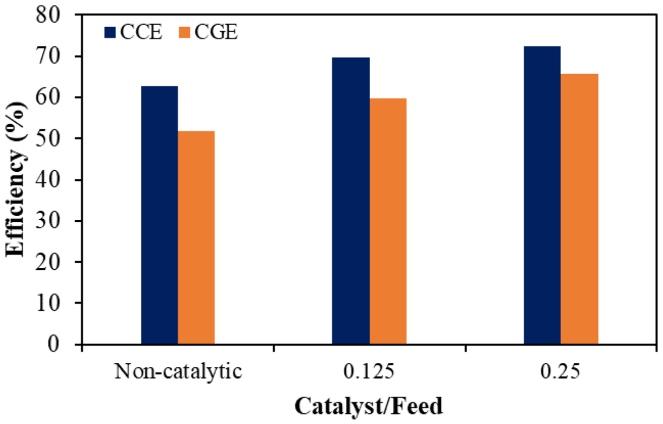

Figure 9: Effect of Catalyst on Gasification Efficiency

Heating value and gasification efficiency of catalytic gasification

The heating values of gasification defined as HHV and LHV are shown in Fig. 8. The highest HHV and LHV were obtained at 19.72 MJ/Nm^3 and 17.70 MJ/Nm^3 at C/F 0.25. Both HHV and LHV are influenced by the concentration of combustible gases, namely CO, H_2, and CH_4. Because the composition of the three gases continues to increase along with C/F, it impacts increasing the heating value.

There are two parameters used as benchmarks for assessing gasification performance: carbon conversion efficiency (CCE) and cold gas efficiency (CGE). The CCE concisely described the process of converting fine coal into syngas Fig. 9 shows the CCE and CGE of the three gasification processes. The gasification process with a C/F of 0.25 has the highest CCE, which reaches 65.61%, while the lowest CCE is obtained in non-catalytic gasification at 51.90%. Carbon conversion efficiency is influenced by the composition of the gas composed by the carbon content. Carbon dioxide is also included in determining CCE.

Cold gas efficiency (CGE) is the ratio of producer gas energy to raw material energy. The highest CGE was obtained at C/F 0.25 of 65.61%. The increase in the catalyst ratio has increased the CGE. An increase in CGE can be associated with a decrease in the amount of CO_2 and an increase in the amount of CO and H_2. The content of volatile matter and fixed carbon also influences CGE [31], [32]. Cold gas efficiency followed the LHV trend because it is a major factor affecting the CGE.

CONCLUSION

In this study, catalytic fine coal gasification was investigated in a fixed bed gasifier using natural bentonite as a catalyst. The temperature has a positive effect on the gasification process by increasing the percentage volume of the syngas composition particularly in H_2. Hydrogen increased from 30.2 vol% to 42.6 vol% from 550 to 750 °C. Increasing the ratio of bentonite to fine coal (C/F) has succeeded in improving percentage volume of H_2 and CO, gasification performance and reducing CO_2. The best syngas concentration was reached at 0.25 C/F and 750 °C (54.3 vol% H2, 26.2 vol% CO, 23.8 vol% CH4, and 3.5 vol% CO_2). The heating value of syngas and gasification efficiency of fine coal are 19.72 MJ/Nm^3 and 72.27%.

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