

Synthetic Gas Production from Fine Coal Gasification Using Low-Cost Catalyst

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ABSTRACT

The utilisation of fine coal waste is still limited, even though its availability is very abundant in the mining industry. This study utilises fine coal by converting it into syngas through catalytic gasification. The gasification process was carried out at a temperature range of 350–550°C for 10–50 minutes using natural zeolite as a catalyst. The syngas composition and quality parameters were evaluated through the H₂/CO ratio, heating value, and gasification efficiency. From the research results, fine coal contained high amounts of carbon and fixed carbon. Temperature is the variable that most influences the gasification process. The addition of zeolite actively increased the CO content in the syngas. The H₂/CO ratio of syngas >1, the highest HHV and LHV 16.15 and 14.46 MJ/Nm³ with the highest carbon conversion efficiency value of 88.85%, made fine coal very suitable to be used as raw material for the gasification process to produce environmentally friendly syngas.

Keywords: syngas, natural zeolite, thermochemical process, energy.

INTRODUCTION

The world's main energy needs are still met by coal. In 2019, the world coal reserves were around 1070 billion tonnes (Singh and Tirkey, 2022). Coal production in Indonesia in 2018 alone has reached 548.6 million tons of coal, and about 21% is used domestically (Baskoro et al., 2021). Fine coal in about 15–20% of coal mines is only stored in ponds (Al-asadi et al., 2020). With the mechanization process and coal mining on a large scale, the amount of fine coal in raw coal continues to increase. The direct use of fine coal will reduce the efficiency of coal utilization and exacerbate air pollution (Wang et al., 2017). Burning low-quality coal causes several environmental problems such as acid rain, air pollution and waste buildup (Munawer, 2018). In commercial settings, this difficult-to-refine coal feed is generally classified as waste, because processing is not economically feasible. Therefore, an

effective and economically viable fine coal processing technique is needed. As one of the technologies for utilizing coal in clean and sustainable energy, coal gasification has received more attention in the last few decades (Yilmaz et al., 2019). Due to the reduced environmental impact and higher efficiency compared to conventional combustion. Gasification produces syngas, which consists mainly of H₂ and CO. The efficiency of the gasification process depends on various aspects, especially the operating temperature of the gasifier. High temperatures are favorable for carbon conversion and tar reduction (Butera et al., 2020; Rosner et al., 2019), but the energy and costs required for the gasification process are also high. At lower temperatures, the problem of low fuel conversion may arise (Saleem et al., 2019). To maintain high efficiency at low temperatures, it is necessary to improve the process by adding a catalyst. The catalyst can lower the gasification temperature, increase the conversion rate, and

increase gas production (Faizal et al., 2021b). Catalysts are used in the gasification of natural minerals, such as olivine (Ma et al., 2019), dolomite (Islam, 2020), and zeolite (Al-asadi et al., 2020; Zhu et al., 2019). The natural catalyst contains alkali metals, such as Na, K, Ca and Mg, which can decompose tar catalytically. Few studies discuss zeolite in the coal gasification process, not even in fine coal. Meanwhile, in biomass and other wastes, gasification using zeolite is still limited (Kislov et al., 2017; Salavati et al., 2019; Valizadeh et al., 2021). Our previous work has used fine coal for gasification using bentonite and purification processes (Faizal et al., 2021a; Faizal et al., 2021b). Therefore, further studies are needed on fine coal in catalytic gasification using zeolite. During gasification, the catalyst is exposed to high temperatures, tar, and sulfur, which can rapidly deactivate the catalyst; therefore it must be durable, abundant, inexpensive, and easily disposed of. Zeolites are advantageous because they are resistant to sulfur and nitrogen, have good thermal stability and have low coke formation. Thus, zeolite is a suitable catalyst that can meet these conditions with low cost and effectiveness in tar removal and H₂S removal during the gasification process. Therefore, this research aimed to produce syngas through catalytic gasification of fine coal using zeolite as a catalyst.

MATERIALS AND METHODS

Feedstocks

Fine coal was collected from Stockpile unit I-IV, PT. PLN (Persero), Bukit Asam Generation Implementation Unit, Tanjung Enim, South Sumatra, Indonesia. Natural zeolite was used as a catalyst in this study with the exact specifications in our previous work (Aprianti et al., 2020). The characteristics of fine coal are known from the proximate and ultimate analysis carried out under three conditions, namely as received (SR), air-dried basis (ADB) and dry basis (DB) according to ASTM D.

Experimental

The gasification process was carried out with variations in temperature (350, 400, 450, 500, and 550°C), reaction time (10, 20, 30, 40, and 50 minutes), and catalyst addition (10 and 20 wt%). The gasification temperature was regulated and monitored via a K-type thermocouple equipped with

a stainless-steel sensor probe and connected to the controller panel. The gasification results pass through a cooling system consisting of a condenser with a diameter of 14.5 cm and a length of 1 m in a horizontal position equipped with a cooling water supply. Fine coal gasification is carried out by collecting data starting from adjusting the ratio between the air entering the reactor and the feedstock used. To start the process, 2 kg of fine coal is fed into the gasifier. The temperature was adjusted as desired from 25°C to 350°C continuing to 550°C. The temperature rise was recorded every 10 minutes. Syngas came out through the gas outlet after going through the cooling and cleaning process. The product gas was accommodated in the gas bag for later analysis of the concentration of its constituents. When the operation time has reached 50 minutes, the data collection was stopped.

Syngas analysis

Gas from the gasification of fine coal was analyzed using gas chromatography (GC Shimadzu-2014). This analysis aimed to determine the composition of the gas produced by gasification. The GC-2014 Shimadzu is equipped with a flame ionization detector (FID) under operating conditions of a Crossbond capillary column (Carbowax Polyethylene glycol) with a length of 30 m, diameter 0.25 mm, film thickness 0.25 μm. The column temperature of 120°C for 7 minutes was then increased to 240°C with a running temperature of 10°C/min, and for 26 minutes, the temperature was constant at 240°C. The detector temperature was 300°C, the injection temperature was 240°C with the Helium carrier gas, the gas flow rate was 3 mL/min, and the gas pressure was 100 kPa. The gas composition under consideration consisted of H₂, CO, CH₄, and CO₂. Through the gas composition, the ratio of combustible gas and non-combustible gas, the ratio of H₂/CO, the value of HHV gas and carbon gas efficiency (CGE) can be determined through several equations that already exist in the literature (Aprianti et al., 2021; Faizal et al., 2021a; Faizal et al., 2021b).

RESULTS AND DISCUSSION

Fine coal characteristics

Proximate and ultimate analyses were conducted to determine the characteristics of fine coal related to its use as fuel. The analysis was

carried out under three conditions, namely as received (AR), air-dried basis (ADB) and dry basis (DB). In the AR-based analysis, the fine coal samples were tested with the initial conditions as the sample conditions in the field. The proximate and ultimate analysis of fine coal is presented in Table 1. On the basis of the proximate analysis results of fine coal, the ash content was 4.79% (AR), total sulfur was 0.45%, and the total calorific value was 19.89 MJ/kg. Fine coal has an ash content of about 5%, which is still acceptable for the conversion process through gasification. High ash content is less favorable for HHV syngas. The environmental parameters regarding gas emissions to be produced are expressed through the sulfur content of the material. From the analysis results, fine coal contained low sulfur <1%, as well as Nitrogen. Thus, the formation of NO_x and SO_x in the released gas will be minimal. The volatile matter content in fine coal explains the ability of the material to evaporate into product gas. Under AR condition, volatile matter fine coal was 33.07%, lower than biomass (Arun et al., 2020; Li et al., 2019; Umar et al., 2021). However, the high carbon content from the analysis of fine coal will contribute to the formation of CO and H_2 in the syngas.

Effect of gasification temperature on syngas composition

Figure 1 shows the distribution concentration on syngas affected by variation temperature. As the temperature rises from 400 to 550°C, H_2 increases from 9.81 to 28.47 mol/L (Figure 1a). The experimental results indicated that the reaction temperature could elevate the H_2 , CO, and CH_4 proportions and relatively reduce the CO_2 concentration. As the

temperature increases, the cracking and reforming reactions intensify again to increase H_2 and CO (Lin and Weng, 2017). The CH_4 concentration increased when the temperature was increased to 450°C and then decreased slowly (Figure 1b). The same thing is also found in the literature that the increase in H_2 during the gasification process is due to steam cracking and tars reforming (Kook et al., 2016; Robinson et al., 2016). The higher temperature favored the production of H_2 due to the endothermic reaction of the steam methane reforming and the char-steam gasification by shifting the chemical equilibrium towards product formation by Le Chatelier's principle (González-Vázquez et al., 2018). The effect of the competing combined reactions showed that the production of H_2 is more dominant than the consumption reaction. The Boudouard reaction increases the carbon conversion, promoting CO formation by consuming CO_2 . Thus, CO_2 continues to fall during the process. CH_4 is formed through the methanation reaction due to the low gasification temperature, which decreases slightly with increasing temperature. The methanation reaction is unfavorable at higher temperatures (Mardani et al., 2021). For irreversible reactions, the reaction rate increased with increasing temperature, but in equilibrium reactions, increasing temperature favored the endothermic reaction and directed the exothermic reaction to the opposite side (Babatabar and Saidi, 2021). Endothermic reactions such as the water-gas reaction, Boudouard, and reforming of steam and methane also increase with increasing temperature. As a result, the concentration of H_2 and CO increased, while the concentration of CH_4 and CO_2 decreased. Furthermore, as temperature rises, the exothermic reactions – such

Table 1. Fine coal characteristics

Parameter		AR	ADB	DB
Inherent moisture	(wt%)	-	13.88	-
Total moisture	(wt%)	30.34	-	-
Ash	(wt%)	4.79	5.92	6.67
Volatile matter	(wt%)	33.07	40.88	47.47
Fixed carbon	(wt%)	31.80	39.32	45.66
S	(wt%)	0.45	0.56	0.65
C	%	58.46	60.70	68.44
H	%	4.69	4.85	5.10
N	%	0.75	0.78	0.89
O	%	12.72	13.16	14.69
HHV	(MJ/kg)	19.89	24.59	28.56

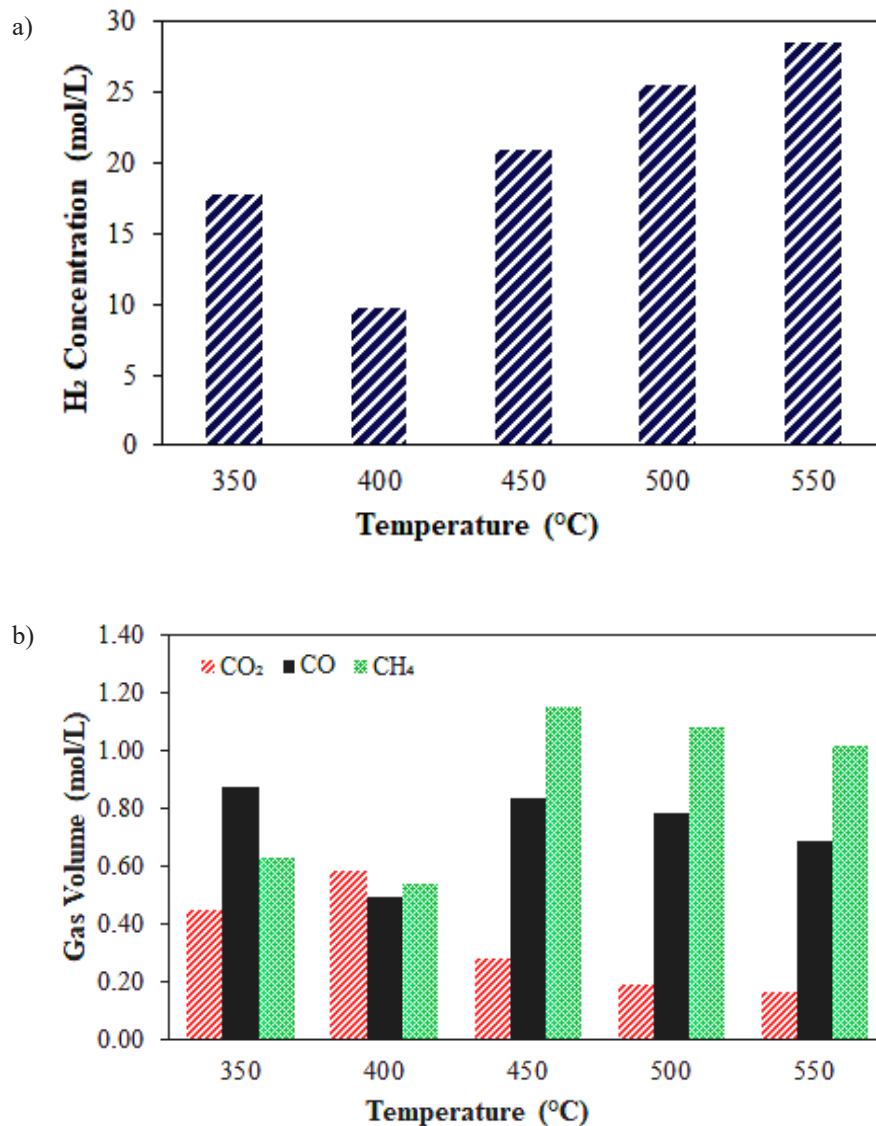


Figure 1. Syngas composition with temperature variation (a) H₂ concentration; (b) Other gases

as water-gas shift and methanation reaction – increase in the opposite direction, resulting in an increase in CO concentration and a decrease in CO₂ and CH₄. The gasification of tar and charcoal is driven by the rise in temperature, which results in greater CO and H₂ production. The other is that the exothermic WGS reaction is inhibited by an increase in temperature, which causes CO₂ and H₂ to react with each other (Xiong et al., 2020).

Effect of reaction time on syngas composition

In this paper, the effect of reaction time on fine coal gasification is studied at 550 °C. The reaction times were set at 10 minutes, 20 minutes, 30 minutes, 40 minutes, and 50 minutes (Figure 2). As shown in Figure 2a, with increasing reaction time from 10 minutes to 40 minutes, H₂ increased

from 3.27 mol/L to 41.94 mol/L, while for CO, it fluctuated slightly (Figure 2b). A longer reaction time causes an increase in the yield of H₂ by increasing the primary reaction that occurs (Rana et al., 2019). This is possible, due to sufficient reagent during this period, as well as the possibility of greater contact between the gasifying agent carrier and the fine coal. Extending the reaction time can increase the WGS reaction, consuming CO and producing H₂ within a certain time limit (Bian et al., 2020). CH₄ increased along with reaction time up to 30 minutes but decreased drastically when the time reached 50 minutes. The formation of CH₄ can be increased with a longer reaction time but is re-consumed through the steam reforming reaction. As the H₂ concentration increases, the CH₄ concentration decreases significantly. Extended reaction time favors H₂

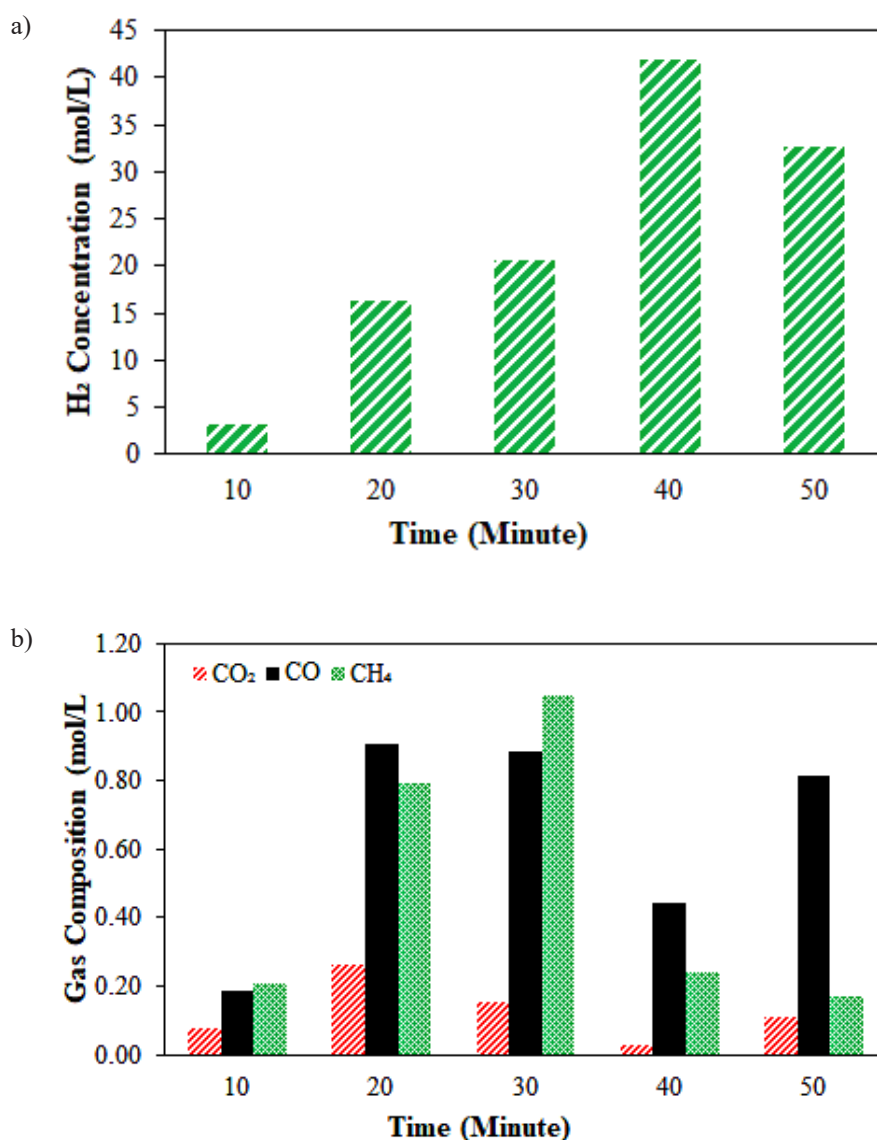


Figure 2. Syngas composition with reaction time variation: a) H₂ concentration, b) Other gasses

production, but not CH₄. The lowest CO₂ content was obtained at 40 minutes of reaction time. Figure 3 shows the effect of temperature and reaction time on the H₂/CO ratio. The H₂/CO ratio of syngas is an important criterion for many industrial applications, including Fischer-Tropsch synthesis, alcohol synthesis, ammonia production, and others. The ratio is set by the dominant WGS reaction in gasification. This reaction is reversible and exothermic in which CO reacts with steam to produce H₂ and CO₂. Because this reaction is exothermic, the equilibrium constant and CO conversion decrease as the operating temperature rises. Therefore, to increase the conversion of the reaction, it must be carried out at a lower temperature. It was observed that the H₂/CO molar ratio increased with increasing gasification temperature. This is due to the fact that at higher gasification temperatures, the rate of increase in H₂ concentration

was greater than the rate of increase in the CO concentration. At high temperatures, the increase in the H₂ concentration continued while the CO concentration began to decrease. In the variation of reaction time, the highest H₂/CO ratio was achieved at 40 minutes, because H₂ was dominantly produced at that time while CO was relatively low.

Effect of addition catalyst on syngas production

The gasification process using zeolite as a catalyst was carried out with the results presented in Figure 4. To see the effectiveness of the catalyst in increasing gas recovery, especially CO, the experiment was carried out at 350°C for 20 minutes using 10 wt% and 20 wt% zeolite. The H₂ content obtained after the catalyst was

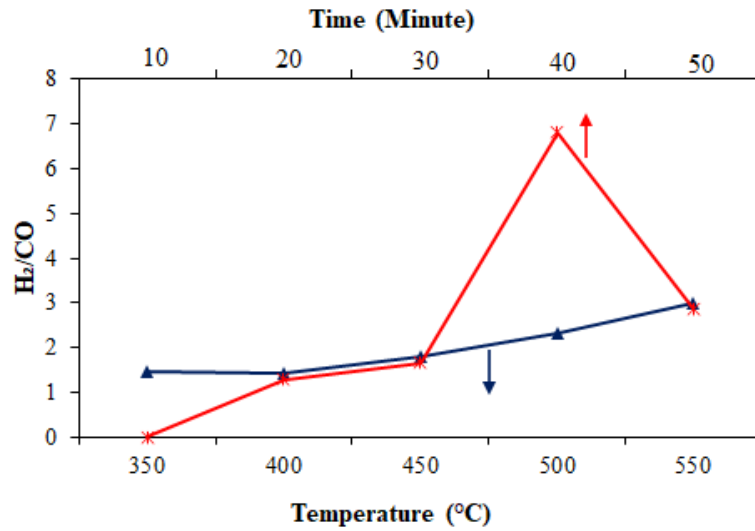


Figure 3. Effect of gasification temperature and time on H₂/CO ratio syngas

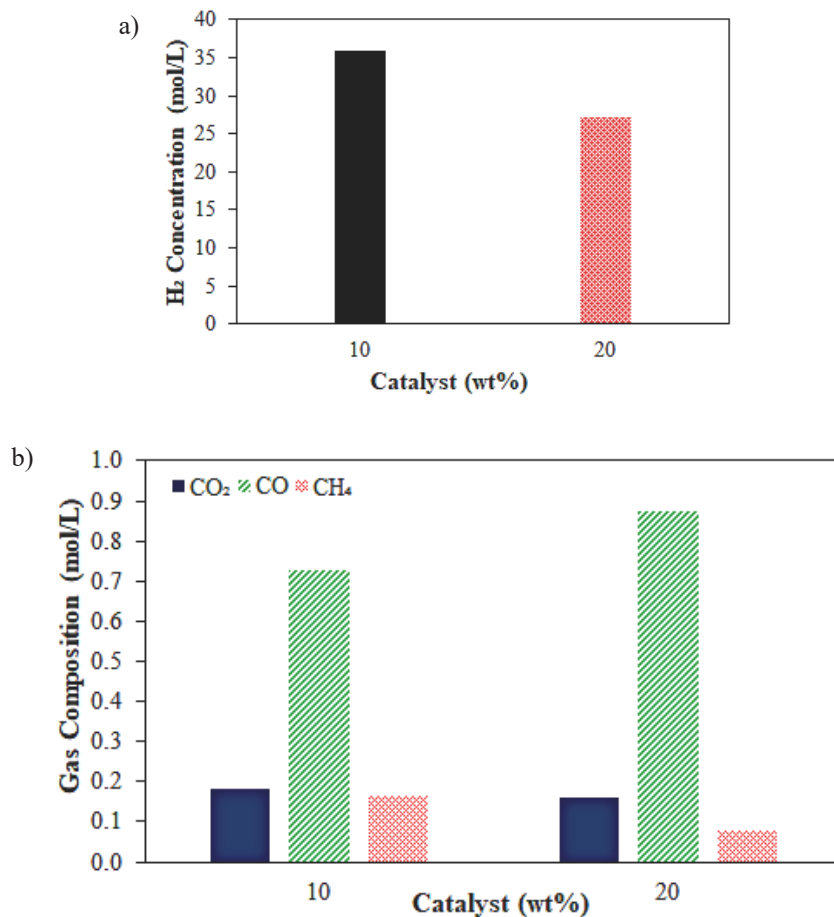


Figure 4. Effect of catalyst addition on syngas composition: a) H₂ concentration, b) Other gasses

applied reached 35.97 mol/L and 27.15 mol/L. The use of zeolites is more effective in increasing the combustible gas, in this case CO, because zeolites have large pores with a wide surface and intra-crystal cavity. Increasing the amount of catalyst from 10 to 20 wt% actually decreased

H₂ and increased CO content. This is following previous studies, where zeolite increased the CO content in syngas (Aprianti et al., 2020). The presence of a zeolite catalyst helps to improve the quality of the syngas by in situ tar reforming and cracking, which breaks down large

molecules (Lalsare et al., 2019). The ratio of H₂/CO syngas obtained after applying the catalyst is 3.53 and 2.22, which makes the syngas eligible for use as fuel and other processes.

Heating value and gasification efficiency

The effect of temperature, reaction time and catalyst addition on the heating value of fine coal gasification is shown in Figure 5. The calorific value increases with increasing temperature. The maximum calorific value of syngas produced is achieved at 500 and 550°C at 16.15 and 16.14 MJ/Nm³, respectively. By increasing the reaction temperature from 400°C to 550°C, the heating value is increased due to the promotion of tar cracking and the hydrocarbon reforming reaction. Increasing the reactivity of the water-gas reaction with an increase in reaction temperature can

accelerate the production of H₂ and CO, which improves the quality of the syngas produced. An increase in reaction temperature will raise the calorific value of the syngas. The quality of the gas produced is determined by a number of factors, including the chemical properties of the feedstock, working conditions, and the gasifying agent. The high calorific value of syngas at higher gasification temperatures is influenced by the increase in moles of CO and H₂ (Ismail and El-Salam, 2017). When time variation is applied, the calorific value obtained is lower. The trend obtained was an increase in calorific value from 10 to 30 minutes but then decreased to 12.33 MJ/Nm³ at 50 minutes. The gasification process, which took about 30 minutes, produces an HHV of about 15 MJ/Nm³. The lower calorific value is caused by H₂, which is higher than CO and CH₄ as the constituent components of the calorific value

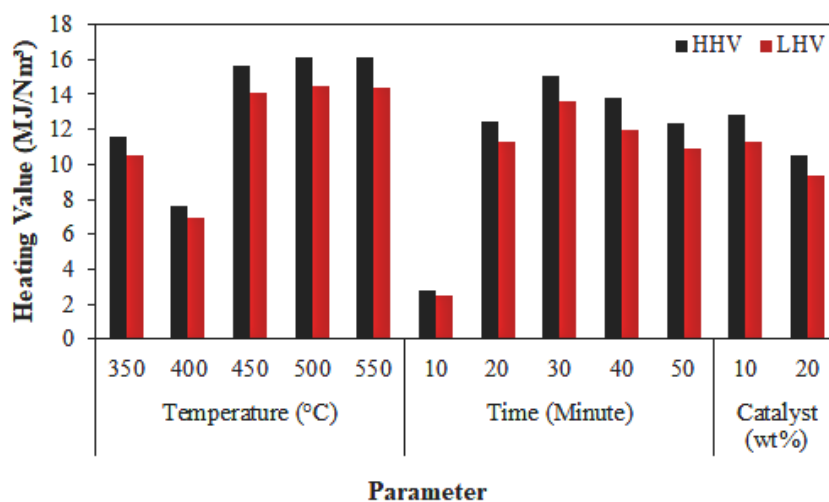


Figure 5. Heating value of syngas

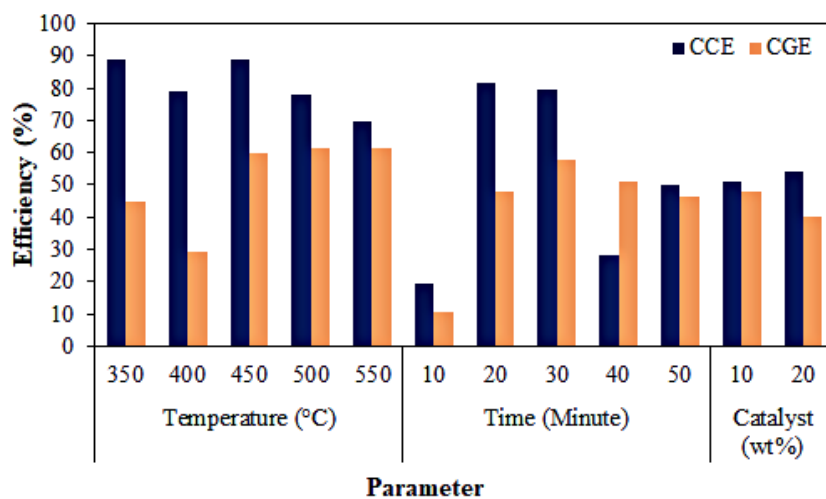


Figure 6. Gasification efficiency of fine coal

calculation. The same thing happens when the catalyst is applied. The advantages of the gasification process are in terms of carbon conversion efficiency (CCE) and cold gas efficiency (CGE) (Figure 6). It can be observed that an increase in temperature causes a decrease in the CCE and CGE of the gaseous products. The CCE of the gasification process was calculated based on the molar flow rates of CH_4 , CO , and CO_2 . Variations in the production of CH_4 , CO , and CO_2 caused the CCE of the syngas to decrease. In addition, the high fixed carbon content in fine coal makes it challenging to gasify, leading to a decrease in CCE and CGE (Huang et al., 2019; Tsalidis et al., 2017). LHV affects the CGE of the syngas so that the obtained trends are similar.

CONCLUSIONS

Fine coal gasification was carried out using zeolite as a catalyst. The results show that the gasification process is influenced by several parameters. The content of H_2 increases along with temperature, while CO_2 exhibits the opposite trend. The reaction time affects the concentration of the gas composition to a certain extent. It was found that the addition of a catalyst from 10% to 20% increased CO over H_2 . Overall, the process can be stated that fine coal waste still has the potential to be used as an environmentally friendly fuel through gasification.

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