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Catalytic Gasification of Fine Coal Waste Using Natural Zeolite to Produce Syngas as Fuel

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ABSTRACT

The valorisation fine coal waste is still very limited in creating energy, especially syngas. This study aims to convert fine coal waste into synthetic gas via gasification using catalyst. Fine coal gasification takes place at 350–750 °C in an updraft gasifier using catalyst of 12.5–25 wt% natural zeolite. The research results show that the addition of zeolite has synergy with increasing temperature. The syngas produced at 750 °C and 12.5 wt% zeolite consisted of 32 vol% H_2, 30.1 vol% CO, 27.7 vol% CH_4 and 5.1 vol% CO_2. The carbon conversion efficiency and high heating value (HHV) of synthetic gas are 88.34% and 18.97 MJ/Nm^3. Fine coal has the potential to be reused as an energy source in the future.

Keywords: fine coal waste, natural zeolite, thermochemical process, hydrogen.

INTRODUCTION

Coal, as one of the primary sources of energy, still plays a crucial role in economic growth and development. In 2020, world coal reserves were recorded at 1,074,108 billion tons, 23.18% of which were owned by the United States (Jiang et al., 2022). Meanwhile, Indonesia reported producing around 548.6 million tons of coal in 2018 and only utilized 21% of them for domestic needs. The total coal in Indonesia is estimated at 140.48 billion tonnes, but only 21.3% is classified as reserves (Baskoro et al., 2021).

In the process of exploration and production of coal, the formation of fine coal is unavoidable. Until now, fine coal in the mining world has been considered a waste with no economic value (Aprianti et al., 2023). In addition to minimal technology in the past, mechanization at each production stage has also produced fine coal in large quantities (Awan et al., 2022). Fine coal is usually discarded and left in stockpiles and sludge ponds, ignoring the fact that this fine coal still has a value to utilize as an energy source. This residual energy source is attracting attention because, if utilized, it will reduce further handling costs, reduce environmental pollution and reduce land for storage.

Coal gasification has always been the foremost and foremost technology in the modern coal chemical industry. Gasification converts carbon-based raw materials into syngas using a gasification medium (air, water vapor, oxygen, or a combination thereof) (Delikonstantis et al., 2019; Sarafraz and Christo, 2020). Through the gasification process, wastes, such as fine coal, agricultural biomass, sawdust, and plastics can be efficiently and effectively converted into fuel (Aprianti et al., 2022a; Hoque et al., 2021; Mansur et al., 2020). In general, the results of gasification consist of H_2, CO, CH_4, CO_2, ash, and tar. Coal gasification is still superior for producing clean and efficient energy compared to conventional processes. Syngas produced from coal gasification has a high calorific value (Solarte-Toro et al., 2018). H_2-rich syngas from coal gasification can produce electricity efficiently (Midilli et al., 2021). In addition, CO_2 emissions are reduced even more through gasification (Lu et al., 2019).

Several studies regarding coal gasification involving catalysts have been carried out by previous researchers. Catalytic steam gasification of coal ash using Ca as a catalyst was carried out by Li et al. (2017). The effect of the catalyst on the gasification process is proven to be recognized from the lower activation energy. Furthermore, using Ca and Na catalysts together with increasing temperature has increased the production of H_2 (Qiu et al., 2018). Yang et al. (2019) studied the catalytic effect of Fe_3O_4 in the steam gasification process of bituminous coal. The iron ore promotes tar reforming and water-gas shift reactions to form hydrogen in the syngas. On the other hand, K_2CO_3 is also used as a catalyst that can stimulate the gasification rate and reduce the gasification temperature (Zhou et al., 2018). Meanwhile, for the zeolite case, the catalyst is still limited for use in gasification. Research on gasification using zeolite has previously been carried out on biomass (Aprianti et al., 2020; Chin et al., 2016; Xie et al., 2019). The results from the studies revealed that CO content increase in the syngas while there is a decreasing phenol content in tar formation. Zeolite as a supported catalyst for Ni showed an increasing H_2 content from 27% to 40.51%. However, the role of zeolite as a catalyst in coal gasification, which synergizes with increasing temperature, needs to be investigated further. Thus, the objectives of this work is to generate synthetic gas from fine coal waste using catalyst of natural zeolite. The effect of temperature and catalyst addition on the synthetic gas quality is evaluated in this work.

MATERIALS AND METHODS

Materials

Fine coal waste was collected from local coal mining in South Sumatra, Indonesia. The characterization of fine coal waste was done in our previous study (Aprianti et al., 2022b). The proximate and ultimate results revealed that fine coal waste with HHV of 28.27 MJ was potentially used as raw material. The fixed carbon, volatile matter, and ash content were 47.67%, 47.59%, and 4.73%, respectively. While the ultimate analysis results consisted of C, H, O, N, and S contents were 74.03%, 5.13%, 14.60%, 1.04%, and 0.48%.

Natural zeolite (<0.5 mm) was purchased from a local area in Jambi Province, Indonesia. The zeolite used in the gasification process was 12.5% and 25% of fine coal weight in the gasifier. The characteristics of the zeolite are known from the analysis of the area, surface morphology, and chemical composition. The area was analyzed using Brunauer-Emmett-Teller (BET). The surface morphology of the catalyst was determined using a Scanning Electron Microscope (SEM JEOL-JSM-6510 LA), while the chemical composition was analyzed using X-ray Fluorescence Spectroscopy (PANalytical Epsilon 3 XLE XRF).

Gasification equipment and procedure

The gasification set-up and procedure were followed the work of Faizal et al. (2021). Fine coal (2 kg) and zeolite were fed into updraft gasifier made of stainless steel. Air as gasification medium was injected from the bottom of reactor. Gasifier coupled with three round electrical elements to reach the desire temperature (350 – 750 °C). The gasification temperature was controlled by two thermocouples connected to control panel. Syngas out of gasifier was subjected to heat exchanger to reduce the temperature and sent into flash drum and filter to separate the gas and tar product. Syngas collected in gas sampling port was analyzed by gas chromatography (GC Perkin Elmer Perkin Elmer Clarus 680). The schematic diagram of gasification process was shown in Figure 1.

RESULTS AND DISCUSSION

Catalyst characteristics

The chemical composition of the zeolite before and after thermal activation in Table 1 shows that silica and alumina are the main compositions of the zeolite. Zeolite consists of more than 70% silica and 14% alumina. Magnesium, calcium, and potassium are also found in zeolite. These elements can swap ions with other metals and nonmetals. At the same time, the remaining elements are impurities that occupy the pores of the zeolite. The number of mineral Al_2O_3 only increased by 0.162% after the activation process. Meanwhile, the amount of SiO_2 decreased by 0.319%. As a

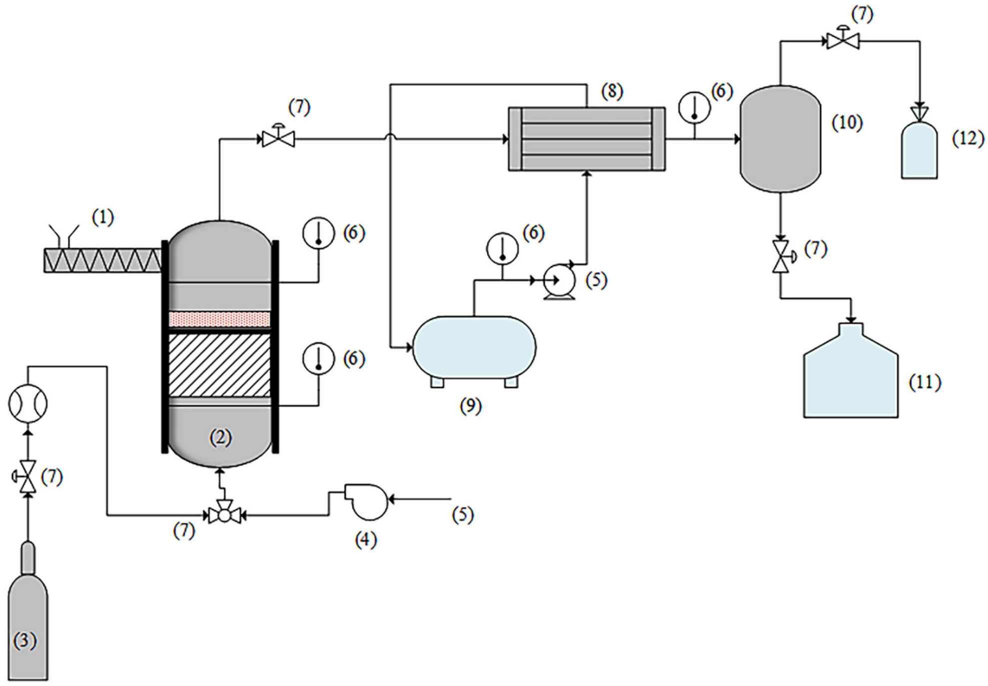

Figure 1. Schematic diagram of fine coal catalytic gasification, (1) feeding hopper; (2) updraft gasifier; (3) N_2; (4) blower; (5) air; (6) temperature controller; (7) valve; (8) heat exchanger; (9) cooling water pond; (10) separator; (11) liquid storage; (12) gas bag

result, the calcination process has no discernible effect on the activation process. Si/Al ratios in thermally activated natural zeolite and natural zeolite were 5.55 and 5.45, respectively. The Si/Al ratio in zeolites did not alter significantly before and after activation. This implies that the natural zeolite was effectively activated without causing structural damage to the zeolite. The high Si/Al ratio in natural zeolite indicates that the dominant zeolite will be clinoptilolite rather than heulandite. Clinoptilolite is present in all natural zeolites, along with certain crystalline impurities, particularly the feldspar minerals calcite, muscovite, and plagioclase, which are common impurities in natural zeolites (Burris and Juenger, 2020).

The results of the SEM analysis in Figure 2 show that some of the smaller particles are spherical in shape with a size of 0.5–1 nm, and most of the particles are large and agglomerated with irregular shapes. Some are porous and feature a layered surface. The surface area of each catalyst before and after activation was obtained by the BET method, which is shown in Table 2. Based on these results, zeolite has surface expansion after thermal activation was carried out. The area of the zeolite becomes 264.344 m^2/g after activation. To some extent, the bigger the specific surface area of the catalyst, the more active sites may be

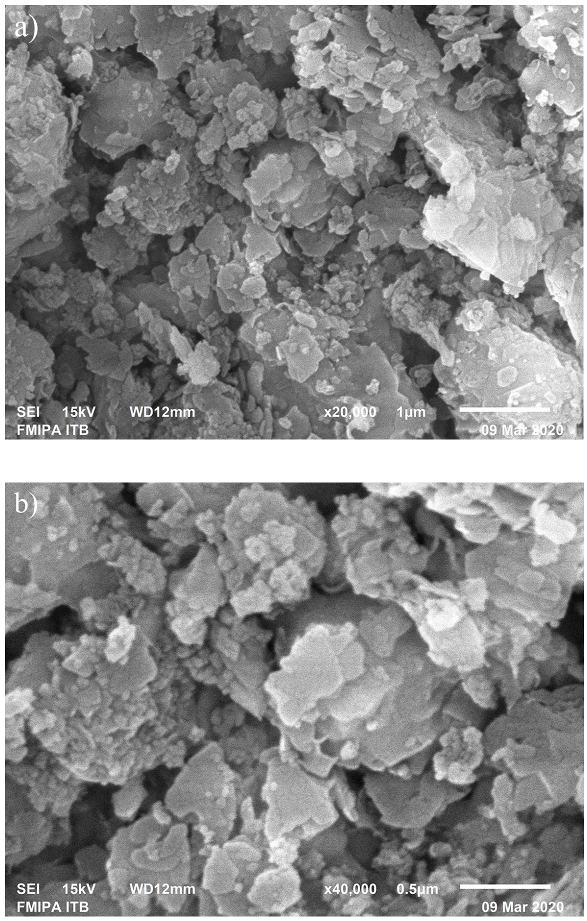

Figure 2. Morphology of natural zeolite at magnification (a) 20,000 times and (b) 40,000 times

given for the reactant molecules, which is helpful for improving the catalyst's catalytic activity (Jiang et al., 2018; Su et al., 2017).

Syngas composition affected by temperature and catalyst

Fine coal is converted to synthesis gas at different gasification temperatures. The gas composition resulting from fine coal gasification, which was evaluated, consisted of H_2, CO, CH_4, and CO_2. From the research results, H_2 increased in the synthesis gas affected by the water-gas shift reaction, which was accelerated by increasing temperature with a final concentration of 42.6 vol% (Figure 3). At 450 °C, the methane generation process in the reduction zone produces 28.1 vol% CH_4 in syngas and reacted again with CO_2 to form H_2. When a result, as the temperature rises from 550 °C to 750 °C, the concentrations of CH_4, CO_2, and CO drop, resulting in a considerable rise in the concentration of H_2. The dry-reforming methane reaction is the name given to the methane reaction. Meanwhile, the CO_2 content at 750 °C is 7.9 vol%.

In catalytic gasification, zeolite increases the volume percentage of CO in the syngas (Figure 4). The H_2 content tends to decrease when the zeolite is applied. The highest CO content was obtained at 750 °C at 33.4 vol%, while H_2 was 30.2 vol%. At 350–450 °C, the CH_4 content increased to 22.1 vol% and 28.6 vol% for 12.5 wt% and 25 wt% zeolites, then when the temperature increased to 550 °C the CH_4 content decreased and increased again until the temperature maximum (750 °C) because CH_4 is formed by the methanation reaction at a higher temperature. The addition of zeolite further increased CO compared to the non-catalytic gasification process and reduced CO_2 volume, which was confirmed by increasing H_2. The acidic nature of the zeolite is a major component of its catalytic activity because it promotes the breaking of the C–C and C–O bonds through the acid site. The number and type of acid sites (Lewis and Brønsted sites) have been reported to influence the cracking process strongly. In general, the CO_2 concentration is lower at higher

Table 1. Chemical composition of natural zeolite (NZ) and natural zeolite thermal activated (NZTA)

Composition	Value (%)	
	NZ	NZTA
MgO	1.140	0.742
Al_2O_3	14.500	14.662
SiO_2	74.733	74.414
P_2O_5	1.369	1.288
SO_3	0.669	0.946
K_2O	2.307	2.461
CaO	3.128	3.235
TiO_2	0.191	0.227
Fe_2O_3	1.618	1.535

Table 2. Surface area of the catalyst

Material	Area (m^2/g)
Natural zeolite (NZ)	124.473
Natural zeolite thermal activated (NZTA)	264.344

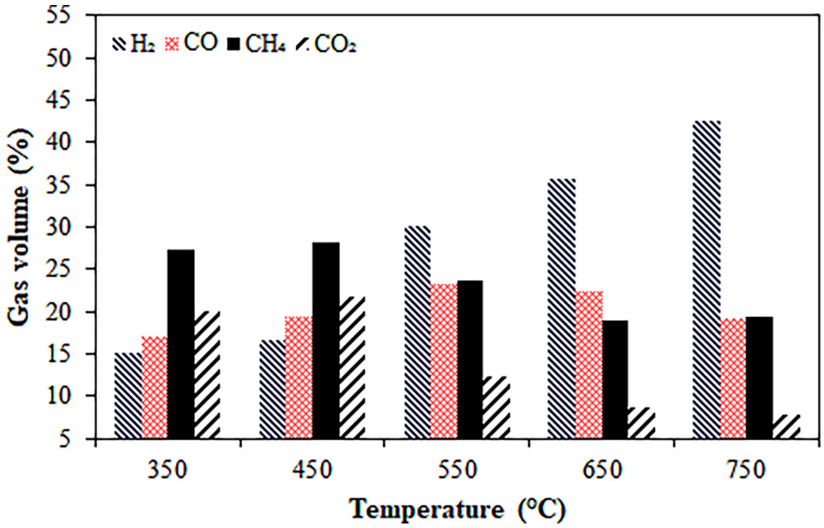

Figure 3. The effect of temperature on the syngas composition resulting from fine coal catalytic gasification

temperatures in each process. According to the thermochemical and thermochemical-catalytic processes, methane concentration is significant at higher temperatures.

Effect of temperature and catalyst on H_2/CO ratio

The ratio of H_2/CO gas from the gasification of fine coal is expressed in Figure 5. The ratio of H_2/CO syngas of fine coal increases with increasing gasification temperature. The increase in H_2 is more significant than the rise in CO, so the H_2/CO ratio increases gradually. If the gasification temperature is relatively low, CO_2 tends to be produced thru the WGSR and methanation reaction (Aydin et al., 2019). Fine coal gasification without a catalyst produces an H_2/CO ratio between 0.89 and 2.23. Apart from being affected by temperature, the H_2/CO ratio is also affected by using a catalyst. The fine coal gasification process produces a higher H_2/CO ratio when the temperature is increased to 750 °C. The fine coal particle size also contributes positively to the flow in the gasifier. Based on Fourier's law, the wider the surface area of a substance, the greater the conductive heat transfer. According to Madadian et al. (2017), the carbon conversion rate can be increased if the raw material has a high conductivity due to its larger surface area.

High heating value of syngas and gasification efficiency

Figure 6 depicts the influence of gasification temperature on the calorific value of syngas on different catalysts. The calorific value of syngas was calculated according to the HHV and LHV equations by Monir et al. (2018). The concentrations of CH_4 and CO contribute more significantly to the

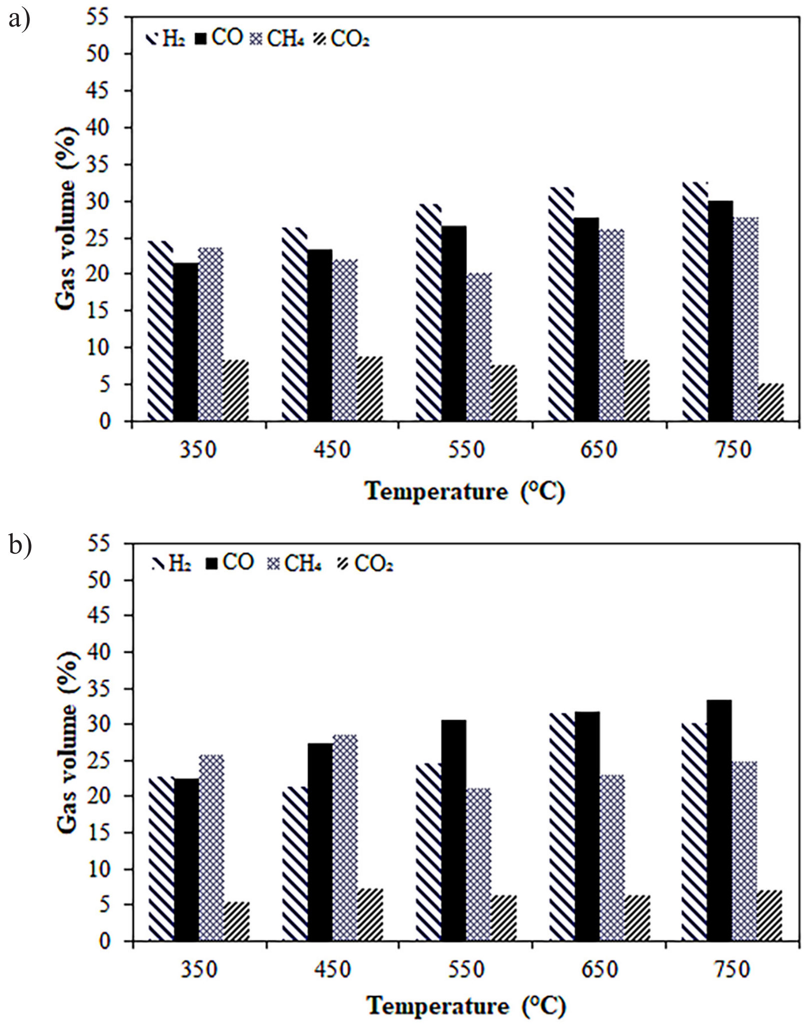

Figure 4. Effect of catalyst on syngas composition from fine coal gasification using 12.5 wt% (a) and 25 wt% (b) zeolite

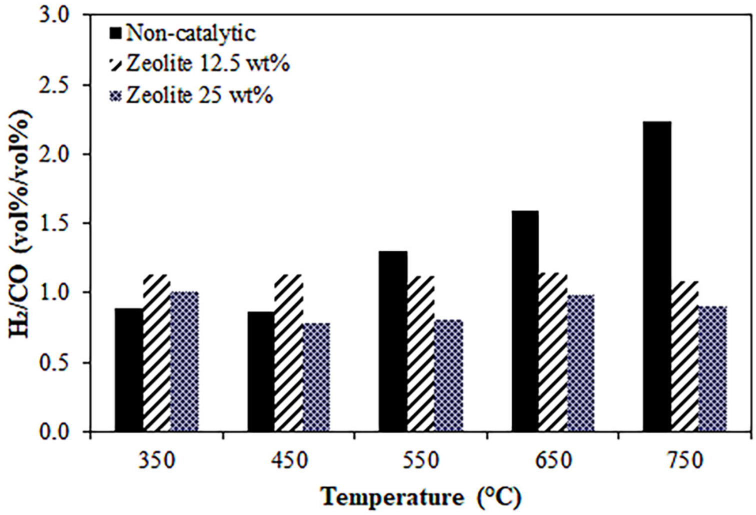

Figure 5. The H_2/CO ratio of syngas from fine coal gasification

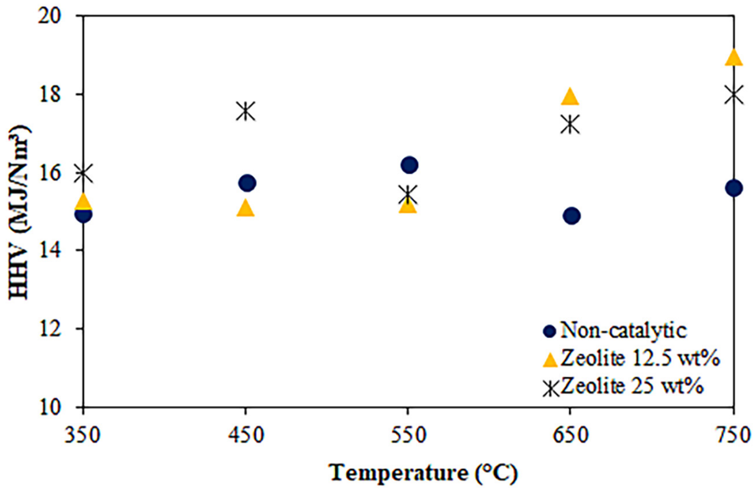

Figure 6. Effect of temperature and catalyst on HHV syngas

heating value of the gas. The system performs well in terms of calorific value, as evidenced by a modest reduction in heat. HHV is affected by combustible gas concentrations, changing due to the application of catalysts, and the heating value is rising. The highest HHV value achieved was 18.97 MJ/Nm^3 at 750 °C. This result is higher than a study conducted by Ma et al. (2019) that used dolomite and olivine catalysts (13.8 and 14.4 MJ/Nm^3).

The performance and efficiency of the gasification process are the basis for assessing its success of the gasification process. The parameters used to measure this are carbon conversion efficiency and cold gas efficiency. Carbon conversion efficiency (CCE) is an important gasification efficiency parameter that describes the fuel conversion process. In non-catalytic fine coal gasification, increasing gasification temperature has reduced carbon conversion (Figure 7a). This is possible because more H_2 is formed compared to CO and CO_2, which are the basis for calculating carbon conversion. Carbon conversion shows stable results ranging from 650 to 750 °C in each gasification process. The highest carbon conversion occurred at 750 °C using a 25 wt% catalysts of 88.34%.

The ratio of synthesis gas energy to raw material energy is known as cold gas efficiency (CGE). The CGE of fine coal gasification increased slowly as the reaction temperature increased (Figure 7b). The highest CGE was obtained at 750 °C using 12.5 wt% zeolites of 63.90%. CGE increases significantly when the zeolite is applied together with increasing temperature. The rise in CGE can be ascribed to a drop in CO_2 levels and an increase in CO

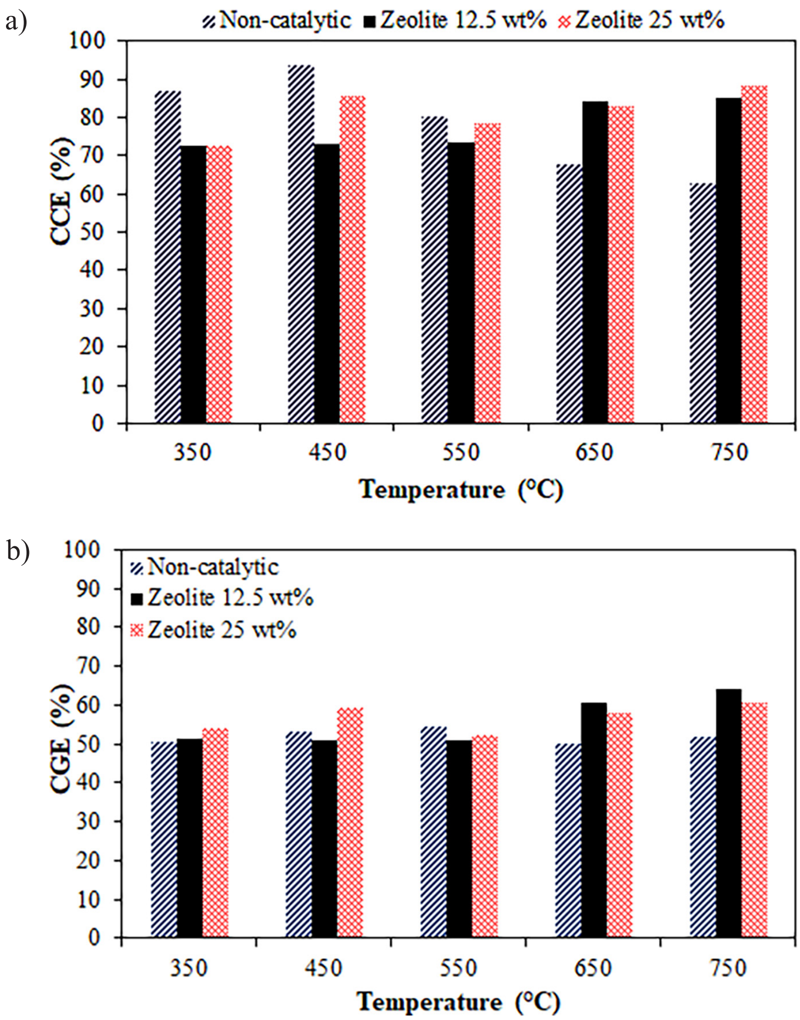

Figure 7. Effect of temperature and catalyst on (a) carbon conversion efficiency and (b) syngas cold gas efficiency

and H_2 levels. CGE is also affected by volatile matter and fixed carbon concentration (Su et al., 2020).

CONCLUSION

Syngas has been successfully created utilizing natural zeolite in an air-medium updraft gasifier from the fine coal catalytic gasification technique. Fine coal has the potential to be used as a raw material for gasification based on its syngas composition, heating value, and process efficiency. The use of a synergistic catalyst in conjunction with an increase in temperature has resulted in an increase in hydrogen concentration. The best performance was achieved using a 12.5 wt% zeolite catalyst with a gas composition of 32.5 vol% H_2, 30.1 vol% CO, 27.7 vol% CH_4, and 5.1 vol% CO_2. The heating value and gasification efficiency were 19.72 MJ/Nm^3 and 72.27%.

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