Enhanced fine coal waste gasification process through Boudouard reaction to produce syngas as energy

by Nabila Aprianti

Enhanced fine coal waste gasification process through Boudouard reaction to produce syngas as energy

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Abstract. Gasification of fine coal waste as waste still produces unavoidable by-products, namely charcoal. The utilization of fine coal and its potential should be advanced to reduce the waste and improve the quality of the synthesis gas produced. The objective of this study is to produce syngas with the addition of fine charcoal to improve the quality of the syngas. Gasification is carried out in a fixed bed reactor at 550-800 °C with different proportions of charcoal. The quality of the syngas was evaluated based on the composition of H2/CO syngas, LHV and gasification efficiency. The results showed that a 50:50 ratio produced a syngas with 45.24 vol% CO2 and 41.38 vol% H2 at 800 °C. Increasing the ratio not only stabilized the heating value gain (~13 MJ/Nm3), but also increased the carbon conversion by > 90%. Alkali and alkaline earth metals (Ca, K, Fe, Mg and Mn) favor the Boudouard reaction to produce CO.

Keyword: gasification, through Boudouard, syngas, energy

1. Introduction

Fine coal is produced during the extraction of coal with low economic value and advanced processing and is categorized as waste in the mining industry. Coal gasification thermally converts coal constituents into gaseous fuels called syngas (CO and H_2), and other byproducts such as tar, a hydrocarbon more than benzene in molecular weight, and solid products (consisting of char and ash). The proportions of the products vary depending on the raw materials and operating conditions [1,2].

Charcoal and ash, which are byproducts of gasification, must be disposed of properly. Charcoal has been used in various processes, for example, in the treatment of dye waste as an absorbent. It is critical to comprehend the link between char's catalytic impact and its consumption (char gasification) throughout the hydrogen generation process from fine char gasification.

Biochar has various distinct qualities as a solid pyrolysis or gasification product, including excellent water-holding capacity and energy density. Currently, charcoal can be used for combustion to provide heat or as an absorption medium and substrate for horticulture [3]. Compared with raw biomass, charcoal is deprived of most volatile components and oxygen, which significantly increases the carbon content. However, there are only a few studies dealing with the use of charcoal to produce high quality syngas.

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Given the low reactivity of char gasification, the addition of a catalyst to enhance the char reaction is necessary when high temperatures are not required, increasing the risk of plant losses. In supporting the catalytic tar cracking process, the char is affected by physical properties such as pore size, surface area, and others [4]. At least three important inherent characteristics, namely the alkali and alkaline earth metal (AAEM), the physicochemical properties (specific surface area, pore size, volume), and aromaticity and surface functional groups in the char, may contribute to the interaction.

The huge specific surface area is unquestionably advantageous for AAEM dispersion or surface functional groups that may impact AAEM catalytic performance via charge transfer [5]. In addition, charcoal has attracted great interest as a substrate or catalyst because it is inexpensive, effectively removes tar, and prevents carbon deposition [6–8]. As far as we are aware, there are few studies dealing with the use of char from gasification to produce syngas. In this work, the main objective of our research is to improve the syngas quality by Boudouard reaction with different feedstock mixing ratios in gasification of fine char waste with bentonite catalyst.

2. Materials and method

2.1 Materials

Fine charcoal with a particle size of 1 mm was obtained from a coal mine in South Sumatra Province, Indonesia. The charcoal was from the gasification process at 750 °C from previous work [9]. The charcoal was collected and sieved to a particle size of approximately 0.5 mm and then analysed for composition by X-ray fluorescence analysis (XRF). The bulk density and BET surface area of the fine charcoal were 1.26 g/cm^3 and 1520.18 m^2/g, respectively. The proximate and ultimate analysis of the fine coal was described in our previous literature [9]. The volatile matter of the fine coal was 41.18%, solid carbon was 41.25%, total moisture was 13.48%, ash content was 4.10%, and heating value (HHV) was 28.18 MJ/kg. The contents of sulphur, carbon, oxygen, nitrogen and hydrogen were 0.42, 64.05, 12.63, 0.90 and 4.43%, respectively.

2.1 Experimental equipments and procedure

The updraft fixed bed gasifier was used with dimensions of (height 670 mm and OD 230 mm). The temperature range used in this study was 550-800 °C at atmospheric pressure. Temperature was measured using K-type thermocouples. A mass flow meter with an ER of 0.4 was used to manage the supply of air as a gasification agent. For each run, 2 kg of feedstock and catalyst were loaded into the gasifier. When the temperature stabilizes, the gasification agent is introduced into the gasifier. At the end of the reaction time, the gasification agent is shut off to gather the gases. The exhaust gas is condensed and collected in a gas bag after passing through a condenser for off-line analysis by gas chromatography (PerkinElmer Clarus 680) to determine the concentration of syngas components (H_2, CO, CO_2, CH_4). Each experiment was repeated at least twice to guarantee the data's dependability.

3. Results and discussion

Gasification charcoal is a residue that consists of unreacted solid carbon and also contains ash. Since some ambient oxidants from the gasifier greatly alter the physicochemical and morphological features of the resultant residue, gasification charcoal differs dramatically from pyrolysis charcoal. Despite of its energy value, char is seen as a waste that must be reduced in order to avoid operational issues, high maintenance costs, and/or expensive gas cleaning systems. The usage of charcoal is required to boost the efficiency of gasification. The addition of charcoal to the gasifier improves the performance of the gasification process. It not only increases the CO content, but also leads to higher LHV and CGE, as in other works [10].

3.1 Effect of Temperature on Gasification of Fine Coal and Char Fine Coal

Gasification of fine coal and charred fine coal was carried out at different temperature ranges (550, 600, 650, 700, 750, and 800 °C) with bentonite catalyst (25 wt%) and the experimental results are shown in Figure 1. It can be seen that in the gasification of fine coal, an increase in H_2 and CO accompanied the increase in temperature. H_2 increased from 36.4% vol to 55.5% vol at 800 °C. The same trend holds for CO. CH_4 first increases at 550-650 °C and then decreases when the concentration reaches 21.2% vol to 16.6% vol. Gasification of fine coal has a positive effect on increasing the concentration of H_2 and CO. The highest concentrations of H_2 and CO were obtained at 800 °C with 34.8% vol. and 29.4% vol. respectively. In the gasification of char, the CO_2 content in the syngas is quite high at 13.2-22 % vol. The CO_2 content decreases with increasing temperature. The trend of CH_4 concentration is different from the CH_4 produced in the fine coal gasification. The CH_4 concentration decreases with increasing temperature from 550 °C to 650 °C and then stabilizes around ~21.8 vol% at 750-850 °C. Temperature is a crucial parameter in the gasification process that has an overall effect on the main gasification reactions, namely the WGS and Boudouard reactions to form H_2 and CO. This follows previous studies investigating the effects of temperature on biochar gasification [7]. Increasing the temperature is beneficial for increasing char and hydrogen production conversion. The Boudourd reaction is an endothermic reaction in which the equilibrium shifts to the right at high temperatures. This indicates an increase in concentration CO.

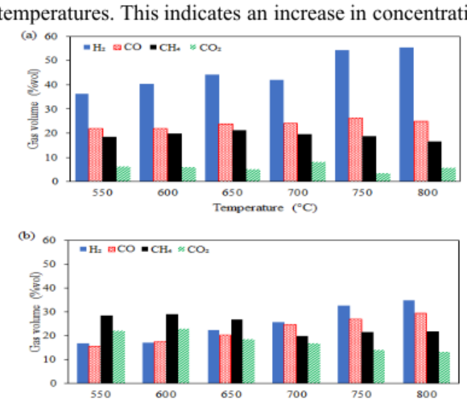

Figure 1. Effect of temperature on syngas concentration in the gasification of fine coal (a) and char fine coal (b)

3.2 Effect of char addition on fine coal catalytic gasification

The reuse of char in the gasification of fine coal has an effect on the syngas concentration, as shown in Figure 2. The H_2 concentration gradually decreased from 55.5% vol to 41.38% vol with increasing char mixing ratio. The CO concentration gradually increased from 24.8% vol to 45.24% vol, while the CH_4 concentration was quite stable at 8.5~9.14% vol. The final CO_2 concentration was 4.26%, suggesting that the gasification event happened on the charcoal's exterior surface. CO Concentration increase. This could be due to the fact that the alkali and alkaline earth metals in the charcoal promote the decomposition of the residual tar in the charcoal. Moreover, CO_2 created in the carbon matrix combines with the charcoal to increase the pores, and light volatiles react with the charcoal at the surface, consuming the carbon matrix [11]. Fine coal is critical to the synergistic behavior of co-gasification, particularly when the quantity of char in the mixture is large. At the same time, the micropores in the semi char created by CO_2 provide more reaction surface for the production of CO [11,12]. These effects, together with the results on charcoal properties presented in Table 1, indicate that the advantages of charcoal are due to two effects. The alkali and alkaline earth metals' catalytic action drives the transformation of the fine char and promotes the cracking reaction/reformation of the hydrocarbons.

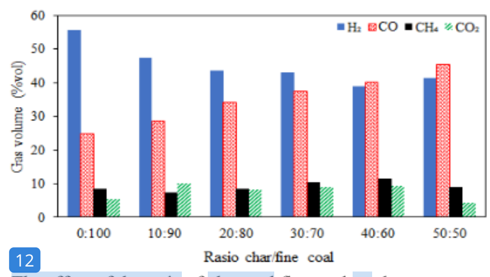

Figure 2. The effect of the ratio of char and fine coal on the syngas concentration

In the gasification of fine coal with charcoal, the Boudouard reaction is stronger. The steam reforming reaction between fine coal and charcoal is due to the relatively large amount of CO_2 and H_2O produced in the pyrolysis phase. In addition, the relatively high amounts of alkali and alkaline earth metals such as Ca and K increase the WGS reaction and the conversion of tar to lighter hydrocarbons [13–15]. Even small amounts of Fe, Mg, and Mn in the fine coal can also increase the reaction rate and gas distribution by breaking down the tar into lighter hydrocarbons. The Boudouard reaction is strongly endothermic and occurs at high temperatures. Because the sole reaction that happens is the Boudouard reaction, coal conversion may be simply calculated from the outgrowth of CO.

Tabel 1. The main composition of char fine coal

Element	Value (%)
Mg	3.179
Al	19.046
Si	33.787
P	3.493
S	13.683
K	1.146

Ca	12.205
Mn	0.189
Fe	9.44

Because of its calorific value and mineral content, reintroduction of charcoal into the gasifier is appealing. Many studies have shown that biochar (often generated from pyrolysis) has a catalytic influence on syngas quality (calorific value, gas output, and/or hydrogen generation) when blended with native feedstocks. The alkali and alkaline earth metal compounds from the gasification feedstocks do not vaporise, but are mostly stored in the char (about 70 – 80 wt%) [16]. These metals facilitate the breakdown of high molecular weight hydrocarbons and tars by supporting numerous gasification reactions [16,17].

Fe and Ca, which are mainly found in biomass and coal, have been shown to be able to reduce NOx precursors and form heavy tar [16]. Moreover, CaO and other ash species (MgO, Al_2O_3, K_2O_3, Fe_2O_3) absorb CO_2, altering thermodynamic circumstances and encouraging alkylbenzene breakdown and H2 production [18–20], and they can even prevent some tars from forming during gasification [21,22]. As the gas combines with the char, the volatiles containing active H and OH radicals are adsorbed on the surface of the char half and chemically react to generate solid products [10,23,24]. The CO_2 in the gas reacts with charcoal or tar which adheres to the surface of the decomposed charcoal [25]. Therefore, it can be assumed that the increase in gas production is due to a change in gas flow in the confined reaction area, recombine volatile components and residual charcoal. Therefore, activated carbon from the gasification process may be a suitable solution to increase the overall gas conversion.

3.3 Effect of Temperature and Char Ratio on H2/CO ratio, LHV and Gasification Efficiency

The H_2/CO ratio describes the direction of syngas utilisation. Figure 3 shows that the H_2/CO ratio in syngas from catalytic gasification of fine coal increases with increasing temperature as it coincides with H_2 production. The H_2/CO ratio briefly decreased to 700 °C, but increased again as the temperature increased to 750 °C and 800 °C. The highest H_2/CO ratio was reached at 800 °C with 2.24. The same trend was observed for char gasification, but the increase was insignificant. A different trend was observed in the syngas produced by gasification of fine coal with the addition of charcoal. In this study, the H_2/CO ratio decreased dramatically from 2.24 to 0.91 when 50% charcoal was used. The rapid increase in the concentration CO with a simultaneous decrease in H_2 results in a low H_2/CO ratio, which is still within acceptable limits for the production of methanol, ammonia, and synthetic fuels via Fischer-Tropsch synthesis [26]. This result is in agreement with the research results of Zaini et al [27], where the H_2/CO ratio decreases with increasing char content in the gasifier.

The H_2/CO ratio is greater than 3, indicating that the high quality syngas generated by char gasification is acceptable for a wide range of applications without requiring any alteration. The hydrogen concentration in char gasification is substantially greater than in biomass gasification because biomass gasification is restricted by thermodynamic equilibrium, hence the H_2 is 40 – 60 vol%. Most crucially, the concentration of CH4 in the gas stays low, which is advantageous for downstream usage. Methane is a contaminant that causes carbon deposition and catalyst degradation in the F–T synthesis process. Low methane concentration is also required when the gas is used in a fuel cell [28]. Moreover, methane reforming is a highly endothermic reaction that requires additional energy, which is not economically advantageous. Therefore, a low methane concentration brings more advantages in the next step.

The lower heating value (LHV) of syngas is calculated based on the concentration of CO, CH_4, and H_2 in the syngas. Figure 4 shows the LHV ratio for gasification of fine coal, charcoal, and fine coal + charcoal. The LHV from gasification of fine coal and char is not too different from the LHV from gasification of fine coal and char, respectively. The LHV varied slightly with gasification of fine coal and charcoal, but increased with increasing temperature. The LHV ranged from 13.3 to 16 MJ/Nm^3 for gasification of fine coal and slightly lower for gasification of char (13-15.3 MJ/Nm^3). The higher LHV results from CO due to higher carbon conversion [29]. The LHV for the gasification combination of fine coal and charcoal is highest at a 50:50 ratio. An increase is seen when char is added at ratios of 10:90, 20:80, and 30:70. However, the increase stagnates when the ratio increases from 30:70 to 50:50. According to Zhang et al. [30], the LHV in the range of 13-15 MJ/Nm^3 is considered high and good for fuel. The LHV should be around 7 MJ/Nm^3 [31].

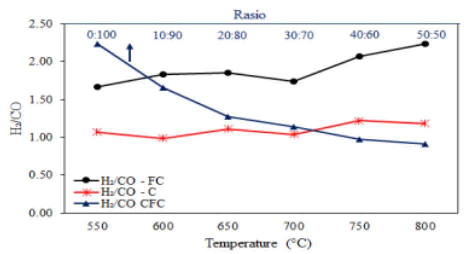

Figure 3. Effect of temperature and ratio of char and fine coal on the ratio of H2/CO

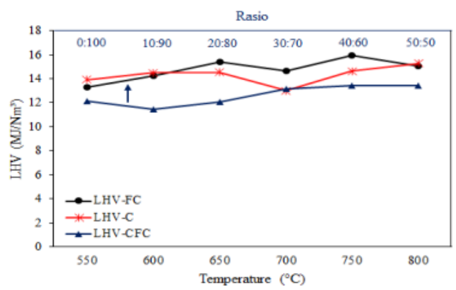

Figure 4. Effect of temperature and ratio of char and fine coal on LHV syngas

The efficiency of the gasification process is viewed from the carbon conversion and cold gas efficiency (CCE and CGE). The CCE of fine coal increases with increasing temperature from 550 - 700 °C. Then, as the temperature increases further to 800 °C, the CCE slowly decreases (Figure 5). Increasing the temperature shifts the reaction towards the product side, which increases the conversion of coal and leads to higher CGE as well as CGE [32,33]. The highest CCE and CGE for fine coal gasification were obtained at 700 °C and 750 °C with 81.19% and 58.97%, respectively. In contrast, for char gasification, the CCE tends to fluctuate and is stable only at 700-800 °C. However, the highest CCE and CGE for char gasification were obtained at 600 °C and 800 °C with 89.19% and 56.65%, respectively. For the gasification of fine coal and char, the CCE increases rapidly with the addition of char. This is due to the high concentration of CO. The ratio of 40:60 between charcoal and fine charcoal gave the highest CCE value with a CCE gain of 95.43%. This result is supported by previous studies that charcoal increases the conversion of carbon to CO. The CGE decreased slightly at first and then stabilized when the charcoal to fine charcoal ratio was 30:70.

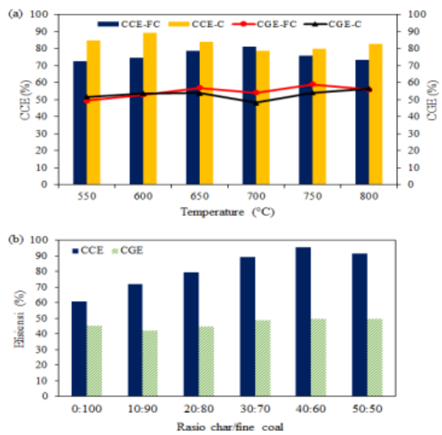

Figure 5. Effect of temperature (a) and ratio of char and fine coal (b) on CCE and CGE syngas

4. Conclusion

Char Fine coal can still be used for gasification, which serves as a raw material and catalyst. From the analytical results, the content of alkali and alkaline earth metals (Ca, K, Fe, Mg and Mn) is favorable for the Boudouard reaction to produce CO. The concentration of CO increases with the charcoal to fine charcoal ratio. The best ratio of 50:50 gives a syngas containing 45.24% vol CO_2 and 41.38% vol H_2 at 800 °C. Increasing the ratio not only stabilizes the heating value gain (~13 MJ/Nm^3), but also increases the carbon conversion by > 90%. The H_2/CO syngas ratio decreased within normal limits with increasing coal content (> 1).

5. Acknowledgements

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