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Purification of Synthetic Gas from Fine Coal Waste Gasification as a Clean Fuel

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ABSTRACT

The presence of CO_2 in the syngas is attracting more attention in terms of reducing the greenhouse gas emissions in its utilisation. The aim of this study was to purify syngas from the CO_2 content of fine coal gasification. Fine coal is gasified with and without absorption using CaO, which is hydrated to $Ca(OH)_2$ in the modified updraft gasifier at 450–700 °C. Apart from investigating the CO_2 absorption process, the gasification process also evaluates the influence of temperature in terms of its synergy with $Ca(OH)_2$. The best conditions for the gasification process are achieved at 700 °C. The content of CO_2 was proven to be well absorbed, which is characterised by a decrease in the CO_2 content and an increase in H_2 in syngas. After the absorption process, the H_2 content obtained increased from 42.6 mole% to 48.8 mole% of H_2 at 700°C. The H_2 ratio also increased after absorption to 2.57 from the previous value of 2.23. The highest absorption efficiency of CO_2 by $Ca(OH)_2$ occurred at 700°C at 50.63%. With an increase in temperature in the gasification process with absorption, the CO_2 content decreased dramatically from 16.9 mole% to 3.9%. $Ca(OH)_2$ has good absorption power at CO_2 at high temperatures.

Keywords: Absorption, CO_2 capture, $Ca(OH)_2$, fine coal, hydration

INTRODUCTION

The use of carbon resources to produce clean fuel is desired to overcome fossil fuel depletion and the resulting environmental problems. In the process of converting a carbon material into energy, minimization of the CO_2 emissions must be considered properly. According to the Ministry of Energy and Mineral Resources of Republic Indonesia (2018), the total amount of coal resources in Indonesia is estimated to reach 105 billion tons, about 21 billion tons of which is reserve; 60% of the reserve coal in Indonesia consists of low rank coal sub-bituminous with the calories content less than 6100 cal/gram. Coal mining in Indonesia, which adopts an open pit mining system, cannot prevent fine coal waste from the production unit and is exposed to the air, watershed and land (Adiansyah et al., 2017; Zhang et al., 2018). This fine material from low-rank coal is very promising for use in the production of environmentally friendly synthetic gases. The synthetic gas from gasification is a mixture of CO, H_2, CH_4, CO_2, and other gases. Syngas can be converted easily into environmentally friendly fuel and used as raw material to produce other useful chemical products.

CO_2 is a by-product of the gasification process, which can contribute to the emissions to the atmosphere. The reduced CO_2 continuously encourages carbon gasification and methane reform and shifts the water gas reaction to the product side so that the syngas is rich in hydrogen (Chen et al., 2020; Chen and Chen, 2020). The heat released from the carbonation process makes up most of the heat supply for the gasification process. The gasification process, producing syngas with low CO_2 emission involves absorbing or capturing

CO_2 in the syngas. Carbon capture and storage are economical and efficient strategies for reducing CO_2 (Ghaemi and Behroozi, 2020; Wu et al., 2020). The Ca-based sorbent is very attractive for use as a CO_2 absorber at high temperatures, apart from being abundant, environmentally friendly, and inexpensive. It is also because of its appropriate kinetics and CO_2 absorption capacity (Soleimanisalim et al., 2016). Calcium oxide plays an essential role in removing CO_2 in syngas through the absorption process in the gasification process. The water-gas shift reaction takes place more dominantly after CO_2 is absorbed so that the purity of syngas, especially H_2, increases (Dou et al., 2016). However, inorganic or dissolved organic calcium compounds derived from limestone are preferred to manufacture synthetic Ca-based absorbers (Soleimanisalim et al., 2017).

The fixation of CO_2 through the production of insoluble carbonate salts, such as through the carbonation of an acidic solution that absorbs CO_2, is another option for capturing CO_2. The aqueous solution of $Ca(OH)_2$ is used as an effective solvent to absorb CO_2 because of its various beneficial features. $CaCO_3$ is formed after CO_2 has been successfully absorbed in the gasifier and can be regenerated (Hafner, Schmid, and Scheffknecht, 2021). Precipitation of $CaCO_3$ from the carbonation of a $Ca(OH)_2$ solution is a widespread reaction, commonly observed in nature. Considering that the cost and stock of the CaO sources in Indonesia are abundant and promising, so the absorption of CO_2 in the gasification process is very relevant for studying. CaO-enhanced gasification is a promising technology for the production of hydrogen-rich synthetic gas (Mostafavi et al., 2016; Sun and Wu, 2019).

The studies on absorption using $Ca(OH)_2$ both as an absorbent and as a catalyst have been carried out by Soomro et al. (2018); Kumar et al. (2019); Kim, Jo, and Kim (2020). The research by Li et al. (2017) found that calcined CaO acts as an absorbent and a catalyst in the water-gas shift reaction in corn stalk gasification. Furthermore, in coal gasification, $Ca(OH)_2$ acts as an active catalyst that decomposes tar to increase the gas yield and hydrogen concentration, which correlates with CO_2 absorption (Shuai et al., 2015; Jiang et al., 2018). Although several studies have been submitted regarding the absorption of CO_2 using CaO or $Ca(OH)_2$ in the gasification process, the research on CO_2 absorption in syngas from the fine coal gasification process using $Ca(OH)_2$ has not been intensively studied by researchers. Therefore, this study aimed to produce syngas with minimal CO_2 in a specific temperature range using hydrated CaO. The gas composition was evaluated to determine the role and synergy between the gasification temperature and the absorbent. The following reactions (R1–4) are occurring within the gasifier in the gasification and purification process:

$$C_{(s)} + CO_{2(g)} \leftrightarrow 2CO_{(s)} \quad \text{(Boudouard reaction)} \tag{R1}$$

$$C_{(s)} + H_2O_{(g)} \leftrightarrow CO_{(g)} + H2_{(g)} \quad \text{(Reforming char reaction)} \tag{R2}$$

$$CO_{(g)} + H_2O_{(g)} \rightarrow CO_{2(g)} + H_{2(g)} \quad \text{(Water-gas shift reaction)} \tag{R3}$$

$$CaO_{(s)} + H_2O_{(g)} \rightarrow Ca(OH)_{2(s)} \quad \text{(CaO hydration)} \tag{R4}$$

$$Ca(OH)_{2(s)} + CO_{2(g)} \rightarrow CaCO_{3(s)} + H_2O_{(g)} \quad \text{(}Ca(OH)_2\text{ carbonation)} \tag{R5}$$

MATERIALS AND METHODS

Raw materials

The fine coal used in this research having the size below 3 mm, was supplied by a coal mining at Sumatera Selatan. The proximate and ultimate analyses of the fine coal are shown in Table 1 in order to investigate the characteristics of fine coal. The gasifier used was a fixed bed type made of stainless steel. Air was used as the gasification agent in this process. The condition inside gasifier was temperature 450°C, 550°C, 650°C and 700°C. The gasifier was isolated to prevent heat release and gas leakage, as well as the two pipes of input stream for gasification agent and output stream for gas product. A flowmeter was installed to measure the flowrate of gasification agent needed. The flow diagram of the gasification process refers to the previous work (Aprianti et al., 2020). CaO was used in the form hydrated by H_2O becoming calcium hydroxide ($Ca(OH)_2$) (R4).

Gasification procedure

The samples were placed into the gasifier and the coal was ignited to 450–700°C, then the mixture of oxygen and air was supplied continuously as the agents of gasification to sustain the gasification process. In this research, the fine coal gasification was carried out under different temperature. The syngas produced then accumulated into gas samplers to be analysed by gas chromatography.

Gas analysis

The syngas produced accumulated into gas sampler bag to analyse by Perkin Elmer Clarus 680 Gas Chromatography equipped flame ionization detector (FID). The success of the gasification process and the quality of syngas are evaluated through the gas ratio and heating value of syngas (Eq. 1–2).

$$HHV_{gas} = = \left((30.18 \times CO) + (30.52 \times H_2) + (95 \times CH_4)\right) \times \times 4.1868\ (MJ/NM^3) \quad (1)$$

$$LHV_{gas} = = \left((30 \times CO) + (25.7 \times H_2) + (85.4 \times CH_4)\right) \times \times 4.2 (MJ/NM^3) \quad (2)$$

RESULTS AND DISCUSSION

Fine coal characteristics by proximate and ultimate analysis

The proximate, ultimate, and calorific analyses in Table 1 provide important insights regarding the chemical properties of fine coal materials. Proximate analysis of fine coal was carried out to identify its composition in moisture, which was later found to be 16.77%. The volatile material of fine coal burning into gas in the nitrogen atmosphere was 40.74%, while the inorganic material in fine coal (ash) was 4.13%. Fine coal is expected to have high gasification efficiency with risks such as clogging, slag, and minor dirt due to low ash. Fine coal contains 38.37% fixed carbon. This result is relatively low compared to raw coal in similar studies (Yan et al., 2021; Vega, Díaz-Faes, and Barriocanal, 2021). The moisture content in fine coal will affect the gasification process. When the water content is high, some of the heat energy will be used to evaporate water in the drying zone. Increasing the volatile matter can result in greater gas conversion yields (Aprianti et al., 2021). Fine coal is mostly composed of minerals. The calorific value of fine coal is relatively high because classified as a sub-bituminous coal type. The high calorific value expected to make the syngas produced also has a high calorific value.

The ultimate fine coal analysis consists of carbon, hydrogen, oxygen, nitrogen and sulphur. The carbon composition in fine coal was 69.04%, which contributes to the CO formation in syngas and a high calorific value. Meanwhile, the H_2 and O_2 content in the material causes a decrease in the calorific value. In fine coal, H_2 was found only 4.78%. N_2 was around 0.97%, because it is an inert gas and is not flammable. The portion of sulphur in this coal type was 0.45%. Oxidized sulphur can later cause air pollution and contribute to the formation of detrimental acid rain. The low nitrogen and sulphur content in fine coal minimise the possibility of NO_x and SO_x formation along the updraft gasification process. Thus, it can be predicted that the resulting syngas is highly desirable for fuel, chemical synthesis and methanol (Mansur et al., 2020).

Effect of reaction temperature on syngas production

Figure 1 shows the synthetic gas after the absorption process at different temperatures. The CO_2 concentration has decreased with increasing gasification temperature from 450°C to 700°C. This situation is due to the fact that the CO_2

Table 1. Proximate and ultimate analyses of sub-bituminous fine coal

Parameter	Unit	Air dry basis
Total moisture	%	16.77
Ash content	%	4.13
Volatile matter	%	40.74
Fixed carbon	%	38.37
Total sulphur	%	0.21
Gross Calorific Value	MJ/kg	21.54
Carbon	%	69.04
Hydrogen	%	4.78
Oxygen	%	13.62
Nitrogen	%	0.97
Sulphur	%	0.45

content in the synthetic gas exiting the gasifier before purification shows an identical trend, where the CO_2 content decreases if the gasification temperature is increased. This can be possible when the CO_2 content in synthetic gas decreases, then the opportunity for the absorption process will easily occur. Hydrogen has increased significantly after the gasification temperature was raised, while methane in syngas tends to decrease. The CO content increases slowly with a range of 20–30 mole% at 450–650°C. When the gasification temperature reaches 700°C, the CO concentration is reduced by 6.5 mole% to 19 mole% at the end of the process. The reaction temperature is an important parameter in the gasification process which significantly affects the equilibrium of the reaction and the resulting syngas (Aprianti et al., 2020).

The effect of CO_2 absorption by $Ca(OH)_2$ on gas composition

The syngas production rate for fine coal gasification with and without absorption is a maximum at a reaction temperature of 700°C. The percentages of H_2 without and with absorption were 42.6 and 48.8 mole%, respectively. This result shows that the CO_2 produced without the absorption process inhibits the chemical reactions shown by R1 and R2, while the absorption of CO_2 using $Ca(OH)_2$ encourages the reaction. $Ca(OH)_2$ has increased the H_2 concentration effectively and decreased the gasification equilibrium temperature (Xiong et al., 2020). The H_2 concentration can be improved with the addition of $Ca(OH)_2$, and the H_2 concentration is more sensitive to temperature by the incorporation of $Ca(OH)_2$.

The carbon monoxide content was observed to increase at 450–550°C, but after passing through this phase, CO decreased when the temperature is increased to 700°C with a final yield of 19 mole%. This is possible because the water-gas shift reaction also increases hydrogen (R3). During the gasification process, methane is consumed through the methane dry reforming reaction. The two reactions are associated with each other and occur in the reduction zone (Kumari and Vairakannu, 2018). Regardless of whether absorption using $Ca(OH)_2$ was applied or not, the amount of methane that fell was not much different (19.5 mole% and 18 mole%) at maximum temperature.

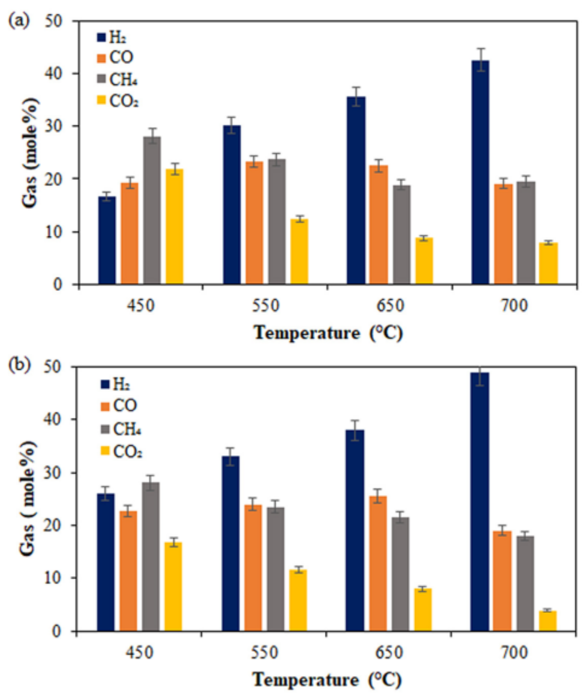

Figure 1. Composition of gas from gasification (a) before and (b) after purification

By adding $Ca(OH)_2$, the higher hydrogen concentration will be obtained under sufficient reaction conditions. In addition, the percentage of gas volume is found to be lower when the gasification process is carried out without the addition of $Ca(OH)_2$. Calcium hydroxide, together with the availability of steam, actively absorbed carbon dioxide. Furthermore, the volume of CO_2 and CO is decreased whilst H_2 is more dominant in syngas by $Ca(OH)_2$. Syngas purification is more likely to occur in gasification after CO_2 has been removed. At the stage between 550–650°C, the addition of $Ca(OH)_2$ only contributes slightly to the decrease in CO_2, but then the performance increases again at 700°C. This is possible because $Ca(OH)_2$ has decreased its ability to absorb CO_2 under these conditions.

CO_2 reduced by $Ca(OH)_2$ is shown in Figure 2. The highest reduction in CO_2 occurs at the initial gasification temperature (450°C). The highest CO_2 content (21.8 mole%) was successfully reduced by 4.9 mole% in this initial state. $Ca(OH)_2$ reduced 22.48% of the total CO_2 in syngas at 450°C. CO_2 remained at the end of the gasification process in a small volume of 3.9 mole%. The absorption of CO_2 in syngas referred to as the carbonation reaction of $Ca(OH)_2$ in gasification. At higher temperatures, the calcination reaction is preferred in the absorption of CO_2. At high temperatures, the $Ca(OH)_2$ particles decompose to become CaO just before CO_2 is absorbed (Li et al., 2015; Yanase, Sasaki, and Kobayashi, 2017). The absorption of CO_2 reduces its percentage volume in the syngas, leading to the formation of H_2, preferably in this state according to the water-gas shift reaction. Hydrogen increases along with temperature and $Ca(OH)_2$ addition. Hydrogen increased by 6.2 mole% while CH_4 slightly decreased after the absorbent applications. These results indicate that the absorption of CO_2 by $Ca(OH)_2$ affects the rate of H_2 production. This situation has been reported in several previous studies (Hwang, Kobayashi, and Kawamoto, 2014; Chen et al., 2017; Lazzarotto et al., 2020). The reaction between $Ca(OH)_2$ and CO_2 formed $CaCO_3$ according to the $Ca(OH)_2$ carbonation reaction (R5).

Gas ratio and heating value of syngas from fine coal

The gas ratio from fine coal gasification is evaluated by the H_2/CO and CG/NCG ratio. The H_2/CO ratio determines the usefulness of the syngas produced (Figure 3). The highest H_2/CO ratio was achieved at 700°C at 2.23. The H_2/CO ratio increases along with temperature. The high H_2/CO ratio is in line with the facts in the previous section that there has been an increase in H_2. As well as the CG/NCG ratio, the CO_2 content that continues to decline coupled with the increase in other combustible gases causes the CG/NCG ratio to continue to increase. In order to see the role of $Ca(OH)_2$ as an absorber, gasification has been carried out at the same temperature range. The results show that $Ca(OH)_2$ has increased the H_2/CO ratio for almost all temperatures except at 650°C. The highest H_2/CO ratio occurred for the gasification process with the absorption of 2.57. The CG/NCG ratio after application of $Ca(OH)_2$ shows a positive effect at each reaction temperature. The CG/NCG ratio before using the absorbent reached 4.83, whereas after using the absorbent it increased to 7.40 at 700°C.

The heating value represents the chemical energy contained in syngas. Humidity affects the heating value of syngas. The pattern of HHV and LHV results obtained between absorption with

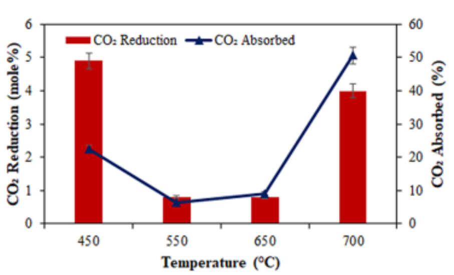

Figure 2. Absorbed CO_2 in syngas by $Ca(OH)_2$ solution

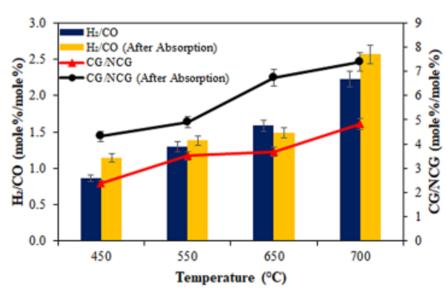

Figure 3. Gas ratio of syngas

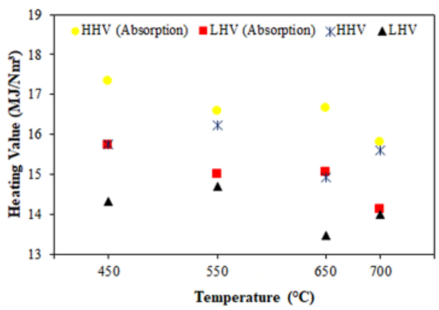

Figure 4. HHV and LHV syngas

and without absorption has a way similar to temperature changes, as illustrated by Figure 4. The highest HHV and LHV were achieved at 450°C with the $Ca(OH)_2$ absorbent of 17.34 MJ/Nm³ and 15.72 MJ/Nm³. CH_4 decreases rapidly when the temperature is increased from 450°C to 700°C, which also has a large effect on the concentration of HHV and LHV syngas. The results show that there has been a significant decrease in HHV from 17.34 to 15.8 MJ/Nm³. Although the H_2 concentration appears to increase over 450–700°C, its heating value is much lower than that of CH_4 so that the HHV and LHV syngas gradually decrease.

CONCLUSIONS

The experimental results demonstrate that the gasification of fine coal with $Ca(OH)_2$ sorbent have a significant impact on syngas producing at the temperature range of 450–700°C. The gas analysis reveal that $Ca(OH)_2$ absorbs the CO_2 content throughout the temperature rise. Additionally, H_2 increased significantly at 700°C from 42.6 mole% to 48.8 mole%. This trend also applies to the H_2/CO ratio, which consistently rises to reach a ratio of 2.57.

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