

CATALYTIC EFFECT OF K_2CO_3 IN STEAM GASIFICATION OF LIGNITE CHAR ON MOLE RATIO OF H_2/CO IN SYNGAS

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ABSTRACT

To fulfill the requirement for synthetic fuel (synfuel) production in the Fischer-Tropsch process, in which syngas fed to the process has a H_2/CO mole ratio approaching 2, gasification of lignite coal is needed. In this research, char particles were prepared by pyrolysis of lignite coal at controlled heating rates to obtain the highest possible surface area for gasification. In the gasification process, char with a surface area of $172.5 \text{ m}^2/\text{g}$ was used, along with the catalyst K_2CO_3 in a fixed bed reactor. In this research, there were variations in the steam/char mass ratio (2.0, 3.0, and 4.0) and in gasification temperature (675°C , 750°C , and 825°C , respectively). Results of this research showed that the highest H_2/CO mole ratio of 2.07 corresponding to the mole ratio of gas yield/carbon of 1.13 was achieved at the gasification temperature of 675°C using the catalyst K_2CO_3 ; the steam/char mass ratio was 2.0. However, at the same gasification conditions—but without a catalyst—the H_2/CO mole ratio and corresponding mole ratio of gas yield/carbon were 3.02 and 0.42, respectively. A finding of this research was that the addition of the catalyst K_2CO_3 to gasification of lignite char adversely reduced the mole ratio of H_2/CO compared to gasification without catalysis. It is suspected that the high composition of mineral ash in coal ash reacted with the K_2CO_3 catalyst, thus causing the Boudouard reaction to compete considerably with the water-gas reaction. The increases in gasification temperature and the steam/carbon ratio lowered the mole ratio of H_2/CO in syngas.

Keywords: Catalytic; Controlled heating rate; K_2CO_3 ; Lignite; Steam

1. INTRODUCTION

To boost domestic energy diversification, the Indonesian government issued Governmental Rule No. 7/2012 to prohibit the export of raw materials from mining (Indonesian Energy and Mineral Resources Department, 2012). The most dominant domestic energy demand in Indonesia is for fuel for transportation. Compared to other alternative fuels, the utilization of synthetic fuel (synfuel) does not require modification of motor engines in vehicles. The physical and combustion properties of synfuel are similar to those of fossil fuels. Synfuel can be produced through lignite coal gasification that yields syngas and Fischer-Tropsch reactions that convert syngas to synfuel. Moreover, gasification technology is cleaner technology compared to direct combustion (DOE, 2011).

Research about Fischer-Tropsch synthesis from syngas with Co/Al_2O_3 , $Co-Re/Al_2O_3$, and $Co-Fe/Al_2O_3$ catalysts showed that syngas with an H_2/CO ratio of 2–2.1 gives the highest rate of conversion to synfuel (Tristantini, 2009a; 2009b). On the other hand, steam gasification of

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lignite coal (high ash content) using a fluidized bed reactor without a controlled pyrolysis process by Handayani, Triantoro, and Diniyati (2013) obtained syngas with an H_2/CO ratio < 2 and 28% carbon conversion at $800^\circ C$; the research also showed that the addition of a K_2CO_3 catalyst decreased the H_2/CO ratio of the syngas product. However, K_2CO_3 catalytic steam gasification of bituminous coal (low ash content) using a fixed bed reactor with no controlled pyrolysis by Wang et al. (2009) obtained a higher H_2/CO ratio compared to gasification without a catalyst. Different characteristics of coal material will give different interaction results to the K_2CO_3 catalyst, which will be discussed further in this paper.

The goal of this research was to identify suitable operating conditions for the gasification of lignite char so that syngas with an H_2/CO mole ratio of approximately 2.0, as well as a high gas yield and carbon conversion rate, would be produced. Pyrolysis of lignite to produce char was conducted separately in a fixed bed reactor prior to gasification of the char to eliminate volatile matter; subsequently, it would expectedly give a high rate of conversion of char to syngas in the gasification step (Quyn et al., 2002). It was found that a controlled heating rate in the pyrolysis step of $3^\circ C/minute$ with a final pyrolysis temperature of $850^\circ C$ gave the largest surface area of $172.5 \text{ m}^2/\text{gram}$ (Chendra, 2014). Gasification with char and steam feeds was applied to enhance H_2 production through a reaction between CO and H_2O (Bell et al., 2011). The addition of the K_2CO_3 catalyst directed the reactions to H_2 synthesis to produce a high H_2 concentration in the syngas product (Satrio et al., 2007). Temperatures during gasification varied (i.e., 675 , 750 , and $825^\circ C$). The mass ratio of steam to char varied as well (i.e., 2.0, 3.0, and 4.0). In this article, findings from the research show the effect of adding the K_2CO_3 catalyst to the gasification of lignite char to produce an H_2/CO syngas ratio of 2.0.

2. METHODOLOGY

2.1. Coal Characteristics

A lignite sample from East Kalimantan in Indonesia was used in this research. Table 1 shows the proximate analysis of the lignite sample.

Table 1 Proximate analysis of lignite coal sample from PT Multi Guna Kalimantan

Analytical Parameter	Result (% adb*)
Moisture in Analysis Sample	14.67
Volatile Matter	40.03
Fixed Carbon	34.50
Ash Content	10.80
Total Sulfur	1.76

*adb: air dried basis

2.2. Experimental Procedure

Coal lumps were dried for 10 hours under ultraviolet light; then, they were crushed into particles no larger than 0.4 mm. After being crushed, coal particles were used to feed the pyrolysis activity in the fixed bed reactor. Pyrolysis at a heating rate of $3^\circ C/minute$ and a final temperature of $850^\circ C$ produced char particles with a high surface area of $172.5 \text{ m}^2/\text{gram}$. Contents of tar and volatile matter in coal evolved from this step. Char particles were thereby ready to be used in the gasification step.

In every run, large char particles (2 g) were mixed with the catalyst K_2CO_3 (10 mass %) during gasification in the fixed bed stainless steel reactor. The sample was supported by quartz wool.

Before the process was initiated, the gasification rig (Figure 1) was purged with a nitrogen flow. Then, the heater was switched on until the gasification reactor reached its operating temperature. As the reactor was being heated, the heater of the steam generator was switched on to heat water to 150°C to generate steam. When the desired temperature was reached, the nitrogen flow was opened and the syringe motor was set to “on” to inject steam. Thus, the gasification reaction was underway. One reaction lasted three hours. Runs under the same operating conditions with no catalyst were also conducted to obtain results for non-catalytic steam gasification.

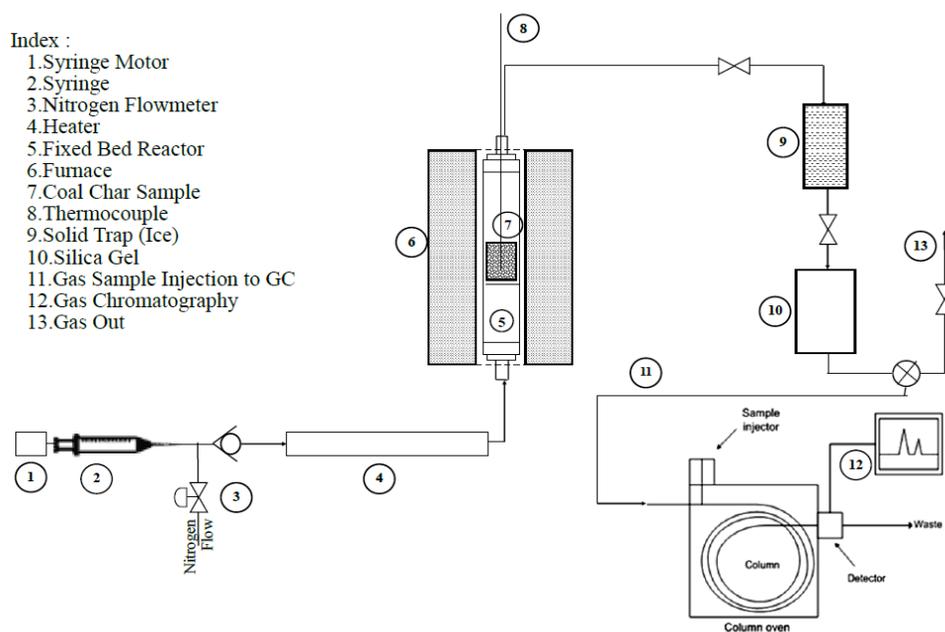


Figure 1 Experimental rig for steam gasification

Syngas output from the reactor flowed through a solid trap, water trap, and sampling port. A syngas sample was taken from the sampling port every 10 minutes using a gas tight syringe for analysis and the gas chromatography-thermal conductivity detector (GC-TCD) technique to acquire data regarding the composition of H_2 , CO , CO_2 , and CH_4 . Based on GC-TCD data, the mole ratio of H_2/CO , syngas yield, and carbon conversion were calculated using a method proposed by Wu et al. (2011).

3. RESULTS AND DISCUSSION

Table 2 shows H_2/CO ratios, carbon conversions, and syngas yields for each condition of steam gasification.

Steam gasification of coal involves several simultaneous thermochemical reactions. Combustion reactions produce CO_2 and H_2O and release thermal energy, which are both needed for gasification reactions. Most important are the water-gas reaction, water-gas shift conversion, Boudouard reaction, steam methane reforming, and methanation reaction (Basu, 2006, Higman, 2008). The existence of H_2 and CO allows the presence of methane steam reforming, from which the methane molecules evolve during coal steam gasification in appreciable amounts in syngas as a result of the reversibility of the reaction (Snoeck, Froment, & Fowles, 2002).

Table 2 H₂/CO ratio, carbon conversion, and syngas yield calculated from experimental samples

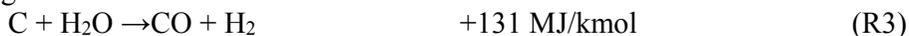
Gasification Temperature (°C)	Steam/Char Mass Ratio	H ₂ /CO Ratio		Total Gas Yield (mole/mole C)	
		Non-Catalytic	Catalytic	Non-Catalytic	Catalytic
675	2.0	3.02	2.07	0.420	1.128
	3.0	2.73	1.92	0.312	0.720
	4.0	2.36	1.80	0.240	0.708
750	2.0	2.26	1.63	0.204	0.636
	3.0	2.04	1.39	0.192	0.384
	4.0	1.98	1.11	0.180	0.276
825	2.0	1.69	1.33	0.192	0.396
	3.0	1.60	1.17	0.168	0.228
	4.0	1.52	1.05	0.156	0.216

Basu (2006, 2010) and Higman (2008) have described the following:

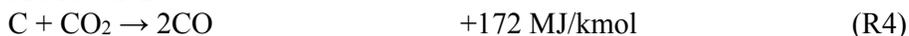
Basic combustion reactions



Water-gas reaction



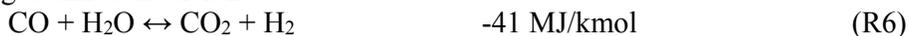
Boudouard reaction



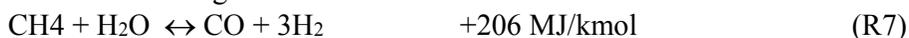
Methanation reaction



Water-gas shift conversion



Methane steam reforming



In terms of the order of reactions (kinetics), reaction R2 has the highest Arrhenius constant in the Arrhenius kinetic constant equation (i.e., 2.2×10^{12}), followed by R6 and R7, both of which have Arrhenius constants of first and second orders of magnitude that are less than that of R2. Carbon reactions in R3, R4, and R5 have orders of magnitude and Arrhenius constants that are much less than that of R1 (Lu & Wang, 2011). The combustion reactions are more than a million times faster than other carbon reactions (R3, R4, R5), but R5 is quite slow in atmospheric pressure compared to the other reactions. Thus, its existence is often ignored (Lu & Wang, 2011).

The use of the catalyst K₂CO₃ not only intensifies the kinetics of reactions R3, R6, and R7 (all of which are gaseous phase reactions), but it also greatly elevates the gasification selectivity toward CO₂, especially at a starting temperature range of 700–750°C (Wang et al., 2009; Wu et al., 2011). It should be noted that high gasification selectivity toward CO₂ fosters a high H₂ production. The kinetics of the Boudouard reaction may be improved in steam gasification of

coal if its ash composition is high (Villacampa et al., 2011) or when gasification is performed at a temperature greater than $700^\circ C$ (Matsumoto et al., 2009). Mendes et al. (2008) found that if char particles are used instead of coal particles, the minimum temperature needed to obtain the high kinetics characteristic of the Boudouard reaction decreases to $600^\circ C$.

3.1. Effect of the Presence of the K_2CO_3 Catalyst on the H_2/CO Ratio in Syngas

Table 2 shows that the addition of the K_2CO_3 catalyst had an adverse effect on the H_2/CO mole ratio in syngas, and syngas from catalytic gasification exhibited a lower H_2/CO ratio than that from non-catalytic gasification. This finding is the same as that for lignite steam gasification conducted by Handayani et al. (2013). However, it is contrary to the results from bituminous steam gasification conducted by Wang et al. (2009), who found that the K_2CO_3 catalyst increases the H_2/CO ratio in the syngas product. These conflicting results indicate that the material characteristics of coal may affect the performance of the K_2CO_3 catalyst. It is postulated that mineral ash may have contributed to the different reaction mechanisms of catalytic steam gasification of lignite and bituminous coal conducted by these researchers.

Leonhardt et al. (1983) found that the ability of alkali cations to catalyze a steam-coal reaction significantly decreases when they react with alumina-silicates, or kaolinite, a mineral dominantly contained in coal ash. The reaction between kaolinite ($Al_2Si_2O_5(OH)_4$) with K_2CO_3 at a temperature greater than $600^\circ C$ will dominantly result in kaliophilite ($KAlSiO_4$) (Kallai & Lapides, 2003), as shown below:



$KAlSiO_4$ obtained from R8 may also deactivate the catalyst, thereby obstructing the formation mechanism of the K-C-O bond that can increase H_2/CO levels (see R9, R10, R11, R12 below) (Wang et al., 2009).

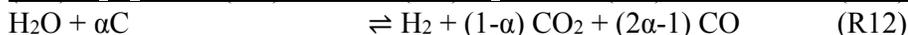
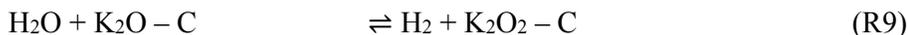
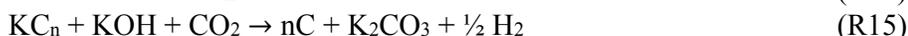


Table 2 also shows that the K_2CO_3 catalyst increases the total gas yield produced. However, because of the interaction between kaolinite and K_2CO_3 , the increase of H_2 production is lower than the increase of CO production. It follows this mechanism in R13, R14, R15 (Matsukata et al., 1988), as follows:



Wang et al. (2010) investigated the addition of $Ca(OH)_2$ to char in catalytic steam gasification to suppress the interaction between the catalyst K_2CO_3 and acid minerals in ash contained in char. They found that the addition of $Ca(OH)_2$ can increase the reactivity of the catalyst compared to the use of raw char. Sharma et al. (2008) investigated the effect of the existence of mineral ash on the kinetics of steam coal gasification and found that catalytic steam gasification of coal with small content ash exhibits nearly four times the kinetics of raw coal gasification. The researchers emphasized the effect of ash presence on catalytic steam gasification, noting that this presence should be in small quantities to achieve high reactivity of the catalyst K_2CO_3 .

However, in the present research, the composition of ash in char particles prepared for catalytic steam gasification was approximately 27%, which possibly affected the reactivity of the catalyst K_2CO_3 . It means that ash material had more exposure to K_2CO_3 particles used in the research as lignite particles have amorphous and more porous structures, thus facilitating contact with the catalyst. Research by Lindstad et al. (2004) conducted by impregnating coke particles with 2–10% K_2CO_3 showed a huge increase of Boudouard kinetics when the reaction occurred above 800°C. The reaction between K_2CO_3 and alumina silicate in ash material seems to exhibit a phenomenon similar to the impregnation of the K_2CO_3 catalyst into carbon material or char, which shifts the role of K_2CO_3 particles from a catalyst for reactions R3, R6, and R7 when they are not attached to char in low ash content to a catalyst for reactions R3, R4, R6, and R7. Therefore, the ability of the catalyst K_2CO_3 to intensify reaction R3 is reduced. This shifting role intensifies the Boudouard reaction, which reduces the mole ratio of H_2/CO in the product.

3.2. Effect of Gasification Temperature on the H_2/CO Ratio in Syngas

Sharma et al. (2008), who conducted a catalytic steam gasification experiment using coal with low ash content at 650°C and 775°C, found that at lower temperatures, gasification favors H_2 production, and at higher temperatures, more CO is produced. This implies that at lower temperatures, gasification of coal is driven predominantly by the water-gas reaction, but at higher temperatures, it is driven by the Boudouard reaction. This result corroborates the finding by Kumar et al. (2009) that at higher temperatures, the Boudouard reaction becomes predominant.

The close interaction between the catalyst K_2CO_3 and ash material allows more heat to be transferred conductively into the char material; thus, the char temperature may be higher than for char with no ash content because of the higher thermal capacity of K_2CO_3 (~3.8 kJ/kg.°C) compared to that of char (~1.5 kJ/kg.°C). Higher char temperatures in the present research were documented, thus intensifying the probability of the Boudouard reaction and increasing the possibility of producing gas with a lower mole ratio for H_2/CO . Table 1 shows that at higher temperatures, the mole ratio of H_2/CO is smaller than at lower temperatures.

3.3. Effect of Steam/Char Mass Ratio on H_2/CO in Syngas

Under present conditions, it is expected that steam gasification is more rapid than CO_2 gasification (Ye et al., 1998). Steam gasification may produce the same number of moles of CO and H_2 . However, the involvement of the water-gas shift reversible reaction changes the composition so that H_2 is present in higher amounts than CO. In their catalytic steam gasification, Luo et al. (2012) found that increasing the steam/carbon ratio causes the composition of H_2 and the mole ratio of H_2/CO in syngas to increase as well. There is an optimum value of 2.10 for the steam/carbon ratio; exceeding this ratio results in lower levels of H_2/CO . The researchers observed that a decline in temperature from an increasing flow of steam may cause the drop in mole ratio. This finding implies that increasing the steam/carbon ratio at a given temperature may increase the mole ratio of H_2/CO in syngas monotonically. In the present research, an opposite result was exhibited. As explained in Section 3.1, in steam catalytic gasification, kinetics of the Boudouard reaction is intensified from the interaction between the catalyst K_2CO_3 and mineral ash, triggering high thermal conductivity spots in char.

At higher steam flow corresponding to a higher steam-to-carbon mass ratio, the heat convection from steam into the char becomes higher due to higher shearing of steam at higher velocity. In turn, the char temperature increases, and exceeding a certain temperature would cause Boudouard reactive kinetics to become appreciable. The formation of CO is governed predominantly by the Boudouard reaction, and the water-gas shift reaction occurring at a higher heat convection rate is attributed to a higher steam-to-carbon ratio. Compositions of H_2 and CO in Figures 2(a) and 2(b) describe these conditions. The concern is emphasized in compositions

of H_2 and CO because they are major components in syngas. In Figure 2(b), at the early stage, the water-gas reaction dominates in gasification at a higher steam/carbon mole ratio, and a lower CO composition results. However, as time proceeds, there is a shifting of the predominant reaction, and the Boudouard reaction takes prominence so that composition of CO is higher than when steam/carbon ratios are lower. The composition of H_2 changes accordingly as the composition of CO changes.

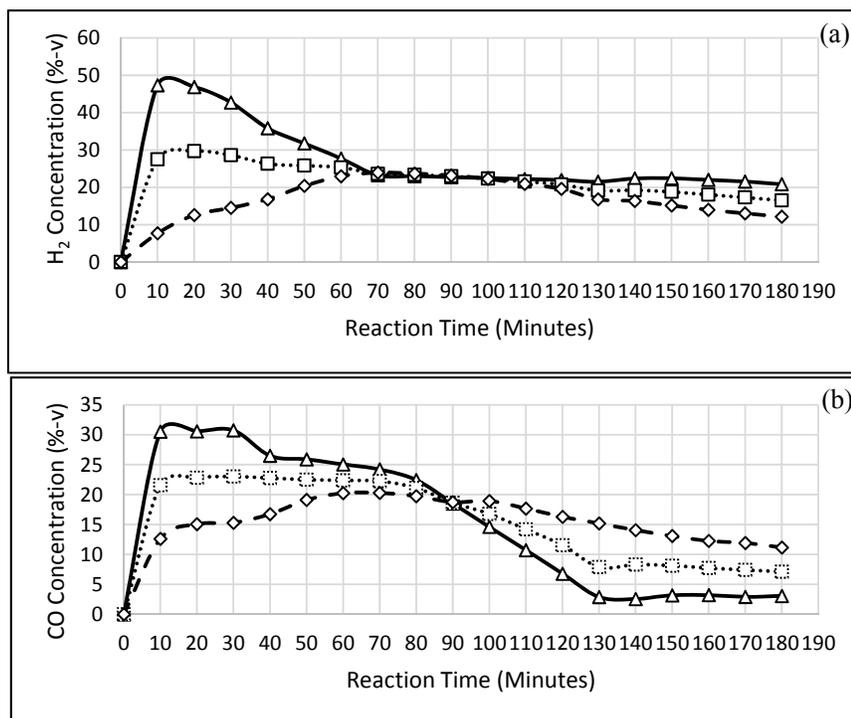


Figure 2 Effect of steam-to-carbon ratio on compositions of H_2 and CO in syngas in catalytic steam gasification at $750^\circ C$ (— \blacktriangle —, S/C = 2.0; \cdots — \square — \cdots , S/C = 3.0, — \diamond —, S/C = 4.0)

4. CONCLUSION

Utilization of Indonesian lignite coal deposits as raw material for syngas/synfuel production may not be feasible through the K_2CO_3 catalytic steam gasification process as the mineral content is too high, thereby interacting with the catalyst. The use of the catalyst may be beneficial if the mineral content in the coal ash is low. In this research, steam gasification using char resulting from controlled pyrolysis (surface area = $172.5 \text{ m}^2/\text{g}$) achieved the highest mole ratio of H_2/CO both without catalysis and with catalysis at a gasification temperature of $675^\circ C$ and a steam/char mass ratio of 2.0. The mole ratios of H_2/CO in syngas obtained were 3.02 and 2.07, respectively, for gasification without catalysis and with catalysis. Increases in gasification temperature and steam/carbon ratio lowered the mole ratio of H_2/CO in syngas.

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