

Comparison of catalytic pyrolysis and gasification of Indonesian low rank coals using lab-scale bubble fluidized-bed reactor

Tae-Jin Kang*, HyeJung Park*, Hueon Namkung**, Li-Hua Xu*, Shumin Fan*, and Hyung-Taek Kim*,†

*Department of Energy Systems Research, Graduate School, Ajou University,
Wonchon-dong, Yeongtong-gu, Suwon 16499, Korea

**Institute of Clean Coal Technology, East China University of Sci. & Tech.,
130 Meilong Rd, Xuhui, Shanghai 200237, China

(Received 4 July 2016 • accepted 27 December 2016)

Abstract—Various methods are used in the coal gasification technology for increasing the efficiency of low rank coal to the level of high rank coal through catalytic gasification. The catalyst used in the catalytic gasification process lowers the activation energy required in the coal gasification reaction. Our purpose was to determine the characteristics of the reaction conditions for producing syngas and the characteristics for comparison catalytic pyrolysis and gasification performance. Among various coals, we used Indonesian low rank coals (Indonesian lignite, MSJ, and Roto South) characterized by a large deposit volume and low cost. Catalytic pyrolysis and gasification experiments were run under the same experimental conditions (reactor type, reaction temperature, catalyst content, and catalyst input method), and the characteristics were compared. Taking the conversion and heating values into consideration, the optimal conditions for catalytic gasification in this study were an H₂O/C mole ratio of 10, temperature of 800 °C, and 10 wt% catalyst impregnation.

Keywords: Low Rank Coal, Fluidized-bed Reactor, Catalytic Gasification, Catalytic Pyrolysis, K₂CO₃

INTRODUCTION

Coal is currently the main energy resource in the world, accounting for the largest percentage among the major energy resources. It maintains more than approximately 40% of the power generation sector, and is expected to continue to maintain a large proportion in the future. According to the 2014 Energy Technology Perspectives (ETP) report by the International Energy Agency (IEA), the demand for energy will likely increase around the world [1]. Clean coal technology (CCT) is an important factor in achieving the goals set for climate change, and this factor requires an increase in the efficiency of coal-fired power plants, as well as carbon capture and storage (CCS) [2].

Generally, coal gasification is carried out at temperatures above 1,000 °C. Low rank coal can be gasified at a lower temperature, but the cold gas efficiency is not high in a fluidized-bed. Although fluidized-bed gasifiers are commonly operated at around 850-950 °C, they suffer from the agglomeration of the bed. Coal gasification has been widely examined at temperature below 800 °C [3-13]. Most low rank coal steam gasification methods have been found to have very low efficiencies and to be impractical [14]. Using a catalyst can significantly enhance the gasification rate at a lower temperature [5-10,15-18]. Thus, the development of coal gasification has several disadvantages, including high reaction temperatures, a large energy

consumption, difficult purification of the products, and stringent requirements for the process equipment. Additionally, it has also been pointed out to cause serious environmental pollution. To resolve some of these issues, a coal catalytic gasification technique was developed to improve the reaction rate and conversion compared to conventional gasification techniques [19]. Recently, catalytic gasification has attracted attention because of its high carbon conversion using less heat energy. Furthermore, catalytic gasification has the important advantage of enhancing the syngas production by decreasing the high pressure and temperature of the gasification condition [20].

Coal gasification can be divided into pyrolysis and the char gasification reaction. Pyrolysis is a chemical decomposition process that can be divided into three parts. The first is the production of an inflammable gas containing hydro-carbons, including hydrogen, methane, and carbon monoxide, by heating organic matter at high temperature, 500-1,000 °C, under anaerobic or low-oxygen conditions. The second product is tar, which contains organic compounds such as acetone-methanol and acetic acid, which are liquids at room temperature. The third product is char, which contains a fraction of fixed carbon, glass, metal, sand, and alkali minerals [21]. Gasification has a very slow reaction rate compared to the pyrolysis process because of the complexity of the reaction due to the internal pore structure caused by the heterogeneous reaction of the solid and gas. For this reason, the reaction rate of gasification determines the overall reaction rate of the coal gasification process [22]. Gasification uses heat, pressure, and steam to convert materials directly into a gas composed primarily of carbon monoxide and hydrogen. Furthermore, losses occur in the reaction due to the saturation of the catalyst in accordance with an increase in the amount of catalyst, and also there are results that suggest that catalyst mixed with excess

†To whom correspondence should be addressed.

E-mail: htkim@ajou.ac.kr

*This paper is reported in the 11th China-Korea Clean Energy Workshop.

Copyright by The Korean Institute of Chemical Engineers.

accumulates in the pores of the coal particles, reducing the contact surface of the porous carbon and limiting the gasification of coal [23]; therefore, varying amount of catalyst was added to compare the characteristics during pyrolysis and gasification according to different temperature and catalyst loading.

Catalytic gasification catalyzes the gasification reaction even at a relatively low temperature and changes the composition of the syngas. In general, because a fluidized-bed gasifier has a relatively short residence time, in order to secure the residence time required for gasification it is used rather than an entrained gasifier.

The purpose of this study was to determine the derived characteristics of the reaction conditions and performance of the catalytic pyrolysis and gasification when producing syngas through a lab-scale bubble fluidized-bed experiment for obtain shorter reaction time and higher carbon conversion using Indonesian three low rank coals. To investigate the utilization of low rank coal, the Indonesian low rank coals Indonesian lignite, MSJ, and Roto South coal were used to analyze the reaction characteristics and syngas compositions of the catalytic pyrolysis and gasification. Various amounts of catalyst were added to compare the characteristics during catalytic pyrolysis and gasification.

EXPERIMENT AND METHODS

1. Sample Preparation

Due to their high reactive characteristic, interest in the use of low rank coals has increased as coal prices have risen as a result of oil price volatility, along with high carbon conversion at low operating temperatures. Indonesian low rank coal is a high grade low rank coal because it has a high moisture content but a low ash content. It is not necessary to carry out elimination process of ash. In addition, there is easy access to the domestic and ensure that the path to import coal. Thus, we selected Indonesian low rank coals with different moisture content, including Indonesian lignite, MSJ, and Roto South. Three Indonesian low rank coals were crushed and sieved at 0.85-1.18 mm. Table 1 shows the results of an analysis of the characteristics of the Indonesian lignite, MSJ, and Roto South Indonesian low rank coals used for catalytic pyrolysis and gasification.

2. Selection of Catalyst

Commonly, a catalyst is used to produce specific syngas products by increasing the selectivity of the reaction [24]. The catalyst reduces the activation energy of the reaction [25]. It is well known that coal and char reactivity are increased by alkaline materials. Among various catalysts, K_2CO_3 has higher solubility than $CaCO_3$ and Na_2CO_3 , K_2CO_3 is regarded as a preferable catalyst, because of its high activity and fair availability. K_2CO_3 interacts with the mineral matter in coal, which makes the recycled use of the catalyst and disposal of the ash difficult [12,13,26]. Therefore, K_2CO_3 is advantageous in catalytic gasification because catalyst deposition is uniform at carbon surface and in the pores of coal. Thus, K_2CO_3 was selected as a catalyst because it has great reactivity in coal gasification.

3. Impregnation of K_2CO_3

K_2CO_3 is known to have excellent reactivity rates in coal gasification. The gasification reaction characteristics may differ according to the mixing method of the catalyst. Therefore, the composition of the syngas was compared through experiments undertaken using the impregnation method for the alkali ion catalyst and coal with the catalyst physically mixed - a mixture of coal and catalyst through physical stirring. Furthermore, the impregnation process was conducted by adding the alkali-based catalysts (K_2CO_3) to water to create an aqueous solution, and then placing the coal in this aqueous solution and stirring for 6 h at 80 °C. Finally, the coal was dried for 24 h at 80 °C. Table 2 shows the results of comparison of the characteristics of the low rank coal and impregnated coal. The results of comparing the raw coal and impregnated coal indicated that the components Mg, Al, Si, Ca, and Fe were reduced equally, whereas the K content was increased. These results show that, K_2CO_3 was 10 wt% impregnated in the coal.

4. Fluidized-bed Reactor and Accessories for Catalytic Pyrolysis and Gasification Experiment

The residence time is important because low rank coal has high moisture content. Thus, the bubble fluidized bed was adapted as the low rank coal gasifier; the residence time could be enough to dry low rank coal. It is well known that the heat transfer of solid-solid in bubble fluidized bed is far superior to that in turbulent and fast transport fluidized beds [27]. The process of catalytic pyrolysis and gasification systems is shown Fig. 1. The lab-scale fluidized-

Table 1. Results of proximate and ultimate analyses of Indonesian lignite, MSJ, and Roto South coals

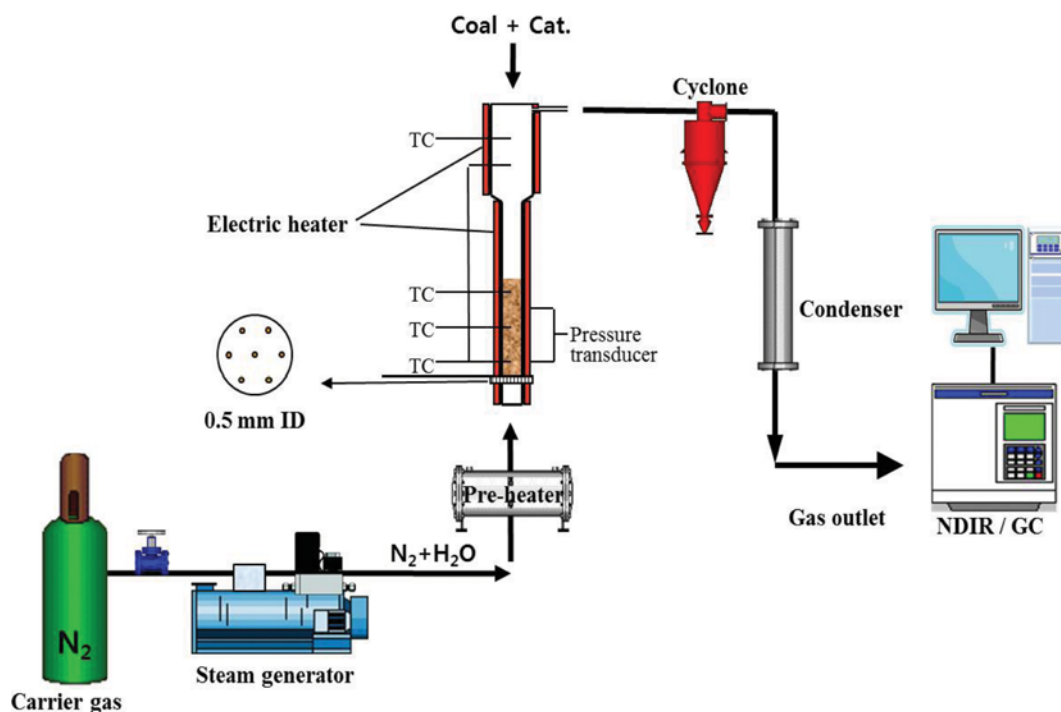
Item	Coal			
		Indonesian lignite	MSJ	Roto South
Proximate analysis wt% (as received basis)	Moisture	32.10	17.16	1.12
	Volatile matter	34.25	38.02	48.75
	Fixed carbon	30.36	41.44	47.95
	Ash	3.30	3.37	2.19
Ultimate analysis wt% (dry ash free basis)	C	52.21 (51.69)	61.72 (59.66)	58.19 (57.16)
	H	4.04 (4.09)	4.96 (4.60)	4.73 (4.25)
	O	42.49 (43.00)	31.50 (34.12)	35.78 (37.42)
	N	0.38 (0.36)	0.87 (0.80)	0.44 (0.37)
	S	0.88 (0.86)	0.95 (0.82)	0.86 (0.80)
HHV, kcal/kg (dry basis)		6,078	6,971	6,154

Note (): K_2CO_3 impregnation coal data

Table 2. Comparison results of low rank coal and K₂CO₃ impregnated coal

	IBC		MSJ		Roto South	
	Raw (%)	Imp.* (%)	Raw (%)	Imp.* (%)	Raw (%)	Imp.* (%)
Na	-	0.04	1.21	0.44	-	0.16
Mg	1.10	0.20	0.87	0.23	1.82	0.32
Al	0.35	0.06	1.58	0.37	3.09	0.51
Si	0.84	0.13	1.32	0.32	5.16	0.93
P	-	-	0.04	0.01	0.03	-
S	3.01	0.61	45.08	8.20	4.60	2.99
Cl	-	0.02	-	-	-	0.03
K	0.24	74.63	0.38	74.7	0.47	79.30
Ca	22.54	10.33	24.96	7.36	27.64	8.73
Ti					1.20	0.18
Mn	1.00	0.19	-	-	0.65	0.07
Fe	70.46	13.72	23.23	7.99	55.05	6.51
Zn	0.41		-	-	-	-
Sr			0.16	0.10	0.24	0.05
Sn			1.09	0.24	-	0.17

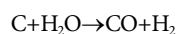
*Imp.: Impregnation

**Fig. 1. Process of pyrolysis/catalytic gasification system.**

bed was made of 310 stainless steel, and coal was fed from the top. It was divided into two zones: the main reactor (ID: 25.4 mm, H: 528 mm) and free board (ID: 50.8 mm, H: 350 mm). In this study, bed material used sand, which is Geldart group B [28]. The lab-scale fluidized bed consisted of components such as the N₂ carrier gas, steam generator, pre-heater, fluidized-bed reactor, main heater, thimble filter, condenser and tar removal device, real-time gas analyzer, data logger, gas chromatograph (GC), and PC.

According to stoichiometry of gasification reaction, steam flow

rate was calculated using carbon content in each sample coal (2 g). In other words, mole ratio of carbon and steam is 1 : 1 for producing syngas.



Therefore, steam requires 1.5 g in a carbon of 1 g. Thus, Indonesian lignite needs 0.255 g of steam, MSJ needs 0.3 g of steam, and Roto South needs 0.2 g of steam at H₂O/C mole ratio 1. The steam generated from the steam generator was injected into the reaction

region, and the N₂ flow rate (7-10 LPM) was set to be 1.5 times U_{mf}. Catalytic pyrolysis and gasification was conducted by simultaneously heating the electric furnace to raise the temperature to reaction temperatures, and injecting the coal sample into the reactor when the reaction temperature was reached. A mixing chamber was installed on the pre-heater to mix the steam and N₂. Because pyrolysis does not require an oxidant, N₂ was used as the carrier gas due to the limitation of carbon. However, the catalytic gasifica-

tion was carried out using steam as the oxidant. The syngas generated through the reaction first passed through the cyclone, in which fine coal and dust were previously removed, and then passed to the condenser to lower the temperature, as well as to trap tar and condensed water to be removed through the outlet. Furthermore, the syngas was passed into the thimble filter, through which the included tar and dust were removed. The composition of the refined syngas was identified through real-time analyzers, and its composition was

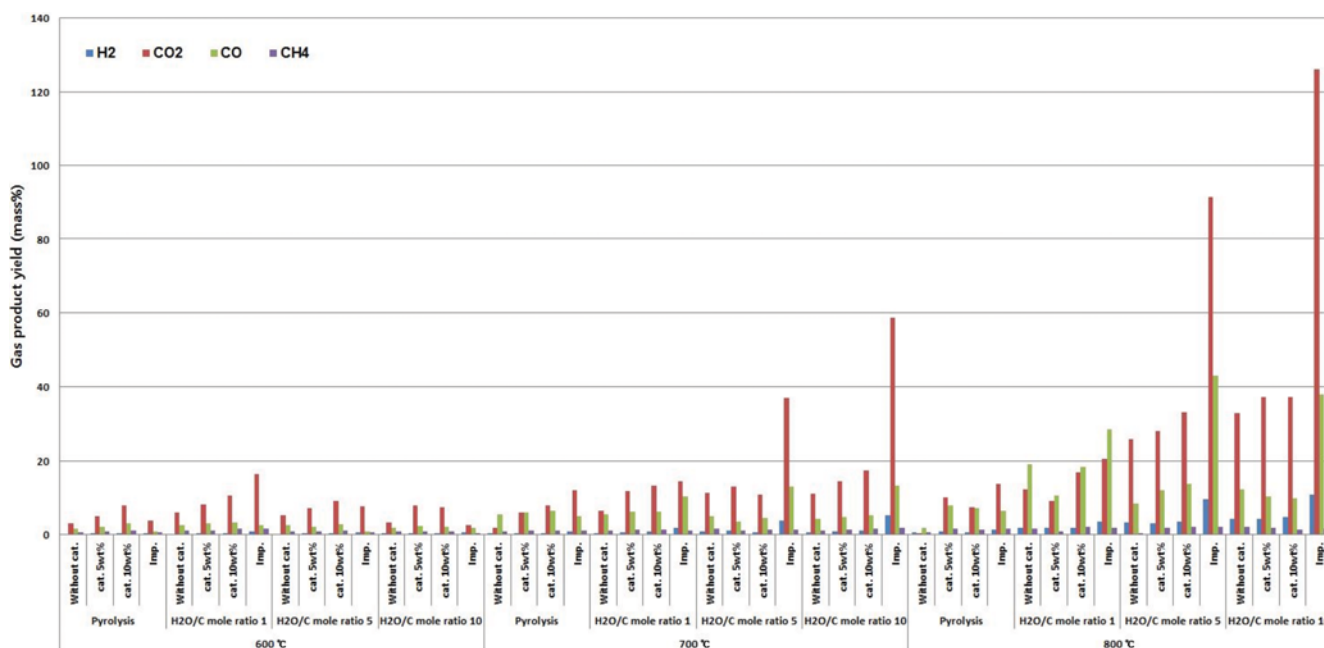


Fig. 2. Comparing effects of reaction temperature on gas product yield for different catalysts in catalytic pyrolysis and gasification using Indonesian lignite.

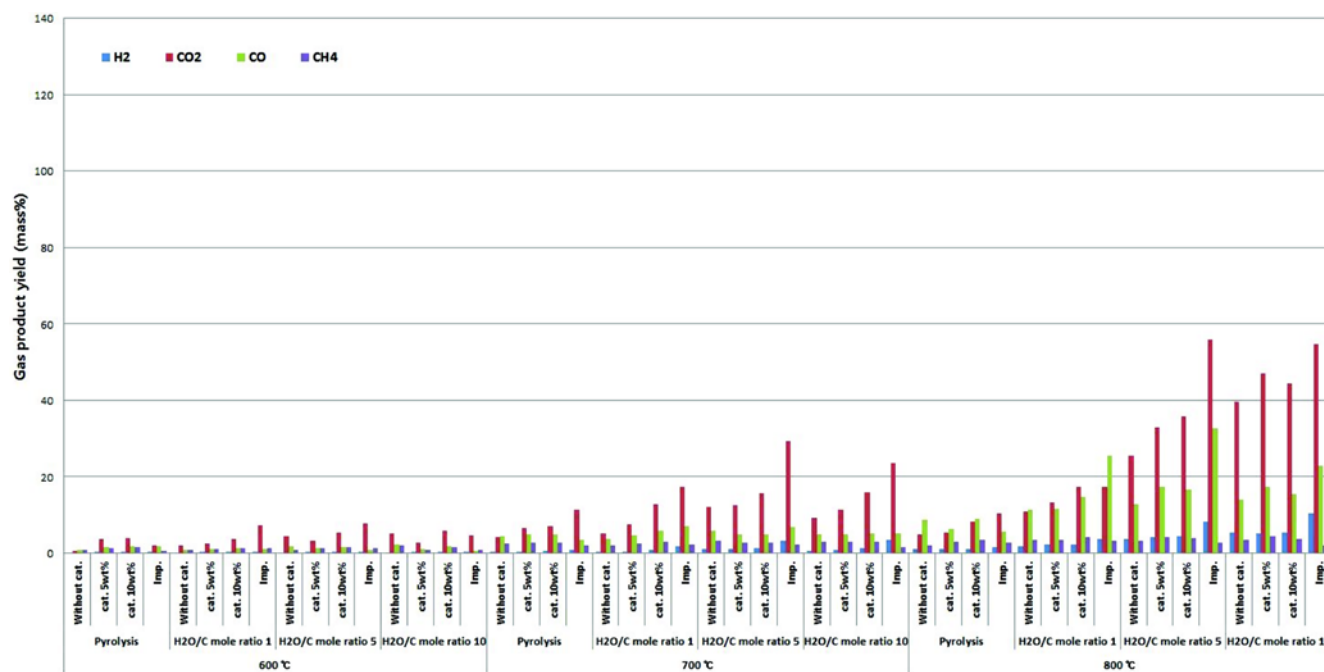


Fig. 3. Comparing effects of reaction temperature on gas product yield for different catalysts in catalytic pyrolysis and gasification using MSJ.

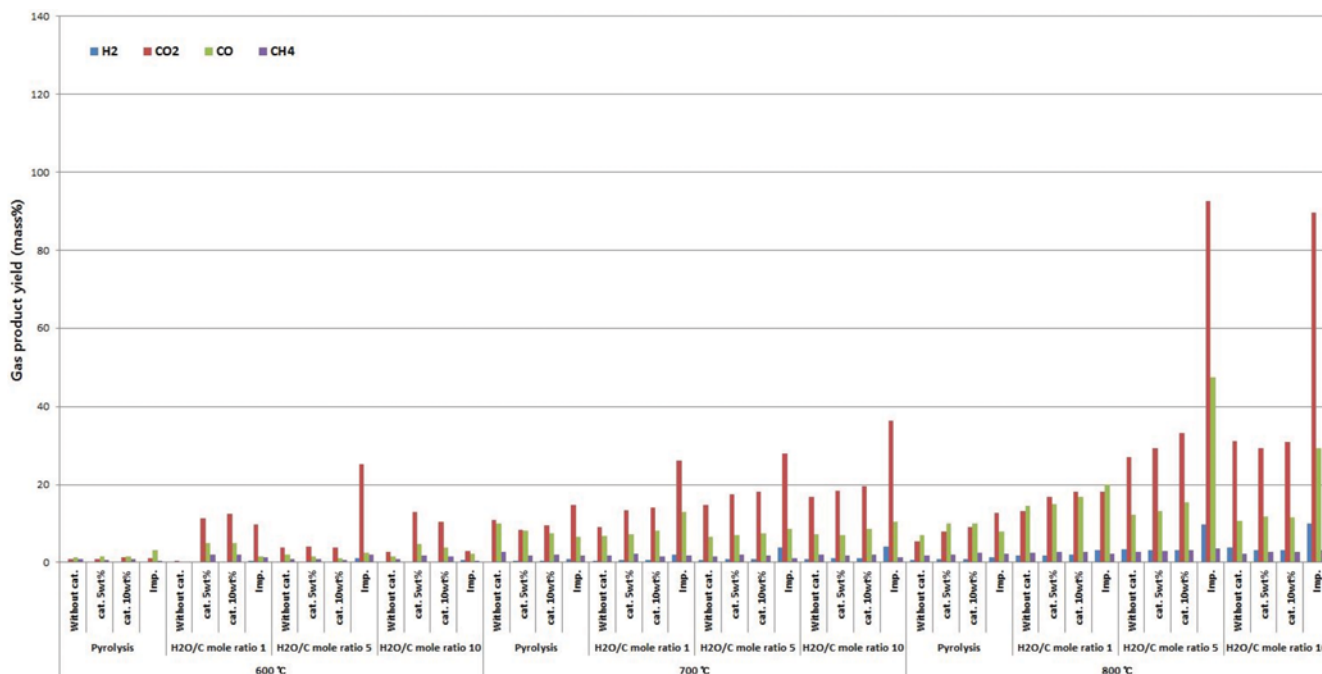


Fig. 4. Comparing effects of reaction temperature on gas product yield for different catalysts in catalytic pyrolysis and gasification using Roto South.

also reconfirmed using the GC.

As experimental parameters, the coal particle size was 0.85–1.18 mm, the coal amount was 2 g, the temperature was set at 600 °C, 700 °C, and 800 °C, and the catalyst (K_2CO_3) injection method was set at 0 wt%, 5 wt%, 10 wt% through physical mixing and 10 wt% using impregnation. In addition, the experiment used H_2O/C mole ratios of to 0, 1, 5, and 10, and the gas velocity was fixed at $U_{mf} \times 1.5$.

RESULTS AND DISCUSSION

1. Comparing Compositions of the Syngas Yield at Different Temperatures

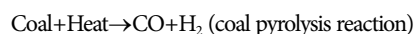
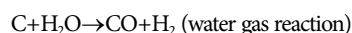
Fig. 2 to Fig. 4 show the compositions of the syngas product yielded at different temperatures. Potassium content in impregnation coal and H/C ratio in experimental sample coals make no difference in Indonesian lignite, MSJ, and Roto South coal. However, considering the O/C ratio in sample coals, the CO_2 and CO produced increased as the O/C ratio increased. From the results, it can be found that syngas composition was affected by O/C ratio, which determines coal rank. The samples, excluding the Roto South coal, were high moisture coals, with a moisture content of over 10 wt%, and contained high levels of volatile matter and fixed carbon. The fuel ratios (fixed carbon %/volatile matter %) of the three samples had values of about one, with a difference of less than 0.2.

The syngas flow rates from catalytic pyrolysis and gasification showed similar trends. In the catalytic pyrolysis and gasification, the syngas production per input coal (g gas/g feed) tended to increase because of the activation of the reaction as the temperature of all the components increased. Therefore, with K_2CO_3 as a catalyst, the effectiveness increases with increasing reaction temperature [29]. Wang et al. [30] reported that no H_2 production was found

at 600 °C in non-catalytic gasification, whereas catalytic gasification produced H_2 at 600 °C. We found the same results in this study. In addition, the H_2 production in this study was higher than that of Wang et al. [30] above 700 °C. Our results showed H_2 production per input coal (g gas/g feed) values of 6.5 for Indonesian lignite, 3.45 for MSJ, and 4.03 for Roto South at 700 °C. Wang et al. [30] reported H_2 production per input coal (g gas/g feed) values of 1.8 at 700 °C and 2.1 at 750 °C for Chinese Huaibei bituminous coal, with 750 °C being the best temperature condition for hydrogen production. Furthermore, as the amount of steam injected increased in the catalytic gasification, the reaction became more active, which led to an increase in the amount of syngas produced. Using K_2CO_3 as the catalyst had an adverse effect on the H_2/CO mole ratio in the syngas, and the syngas produced through catalytic-gasification had a higher H_2/CO mole ratio than that from non-catalytic gasification. Thus, the K_2CO_3 catalyst increased the H_2/CO ratio in the production of syngas [19].

In both catalytic pyrolysis and gasification, the increases in H_2 and CO were much larger than those of CO_2 and CH_4 as the temperature increased. This means that in commercial gasification, H_2 and CO increases are also expected due to the gasification reaction as the temperature increases, but it is believed that the pyrolysis activation will have clear influences on these H_2 and CO increases.

In the catalytic gasification, the sources of the H_2 and CO production according to the temperature increase can be mainly summarized as follows [31]:



At a higher temperature, more CO is produced. This implies that

at a higher temperature, the production is driven by the Boudouard reaction [32]. This result corroborates the finding by Kumar et al. [33] that at a higher temperature the Boudouard reaction becomes predominant. The H_2 and CO increases when using K_2CO_3 as a catalyst are larger at relatively high temperatures, just as in the results of other studies [29,34]. For the H_2 and CO, the slope of the catalytic gasification is larger than that of the catalytic pyrolysis, because the water gas reaction ($C+H_2O\rightarrow CO+H_2$) is enhanced by a temperature increase. This implies that at lower temperatures catalytic gasification is driven predominantly by the water gas reaction, and at higher temperatures it is driven by the Boudouard reaction [33].

CH_4 shows little change with the reaction temperature, and the amount of CH_4 from catalytic pyrolysis and gasification is found to be lower than that from non-catalytic pyrolysis and gasification. This may be due to the decomposition of CH_4 by the catalyst [34].

Furthermore, the production of syngas increases as a result of the effect of the catalyst, whereas the production of H_2 becomes much better when using the impregnation method instead of a physical mixing method. About 1 L of hydrogen was produced at 800 °C, with an H_2O/C mole ratio of 10 and a 10 wt% catalyst physical mixing condition. However, particularly under the condition of the H_2O/C mole ratio 5 or 10 and a reaction temperature of 800 °C, at least 2 L of H_2 could be obtained using 2 g of impregnated coal. Table 3 shows the quantities of H_2 produced at the different temperatures and H_2O/C mole ratios using the impregnated coal. From Table 3, it was found that catalytic gasification conditions of 800 °C and an H_2O/C mole ratio larger than five using impregnated coal could produce about 2 L of hydrogen. As shown in Fig. 2 to Fig. 5, relatively linear changes occurred in both the catalytic pyrolysis and gasification to the H_2O/C mole ratio and temperature. Table 4 shows the yields of the produced syngas in relation to the optimum conditions for the catalytic pyrolysis and gasification.

2. Comparing Carbon Conversion and Heating Value in Catalytic Pyrolysis and Gasification

Fig. 5 to Fig. 7 show the carbon conversion and heating values in

Table 3. Quantities of H_2 produced by impregnation method (unit: ml)

	H_2O/C mole ratio			
	0	1	5	10
Indonesian lignite				
600 °C	82	171	121	121
700 °C	187	388	837	1161
800 °C	277	806	2,162	2,439
MSJ				
600 °C	51	67	68	73
700 °C	197	394	711	774
800 °C	329	815	1,859	2,321
Roto South				
600 °C	61	74	268	151
700 °C	209	431	855	904
800 °C	292	714	2,171	2,217

0, 1, 5, and 10 are H_2O/C mole ratios

Table 4. Yields of products gas for catalytic pyrolysis and gasification in bubble fluidized-bed at 800 °C

Catalyst (wt%)	H_2O/C mole ratio	Gas Composition	Product yield (ml/s)		
			Indonesian lignite	MSJ	Roto South
Impregnated 10 wt% K_2CO_3	0	H_2	1.97	1.96	1.64
		CO_2	1.22	1.00	1.24
		CO	0.79	0.82	0.95
		CH_4	0.33	0.45	0.52
	1	H_2	2.78	2.28	2.17
		CO_2	1.43	0.91	1.23
		CO	0.94	0.89	0.87
		CH_4	0.44	0.80	0.53
	5	H_2	9.32	5.46	5.94
		CO_2	5.17	2.91	4.44
		CO	1.97	1.07	1.51
		CH_4	0.45	0.52	0.68
10	H_2	9.75	9.32	7.34	
	CO_2	6.83	2.23	4.05	
	CO	1.34	1.45	1.16	
	CH_4	0.33	0.41	0.66	

relation to the reaction temperature and catalyst content in catalytic pyrolysis and gasification. From these figures' results, we found that the carbon conversion was less than 20% in pyrolysis; however, catalytic gasification provided 90%. It is well known that the gasification process has presence of oxygen but pyrolysis is absence of oxygen, since pyrolysis could be limit of carbon.

As shown in Fig. 5 to Fig. 7, the carbon conversion and produced syngas heating value increased as the temperature and catalyst feed increased. This result corroborates the finding by Handayani et al. [19] that K_2CO_3 could increase the heating value. As the temperature increased, the heating value tended to increase because of an increase in the amount of produced syngas through the activation of the water gas shift reaction ($CO+H_2O\leftrightarrow CO_2+H_2$, $\Delta H=-42$ KJ/mol) and activation of pyrolysis. The heating value increased with increasing reaction temperature in the catalytic gasification, because the increase in the steam-char gasification rate was larger than the decomposition rate of the hydrocarbons [35]. The carbon conversion in this study was higher than those found by other researchers. Handayani et al. [19] reported that by using Indonesian low rank coal in the fluidized-bed at 800 °C condition, the carbon conversion was less than 60%. However, Hauserman et al. (1992) reported that using K_2CO_3 in the fluidized-bed at 700 °C condition, the carbon conversion was less than 80%. This was because the coal particle size and H_2O/C mole ratio were different from other feed utilized in the fluidized-bed in other studies.

Taking the conversion and heating values into consideration, the optimal conditions for catalytic gasification in this study were an H_2O/C mole ratio of 10, temperature of 800 °C, and 10 wt% catalyst impregnation. Furthermore, because heat was applied by using a furnace, the cold gas efficiency was higher than the conversion, with a value of about 80% under the optimum conditions

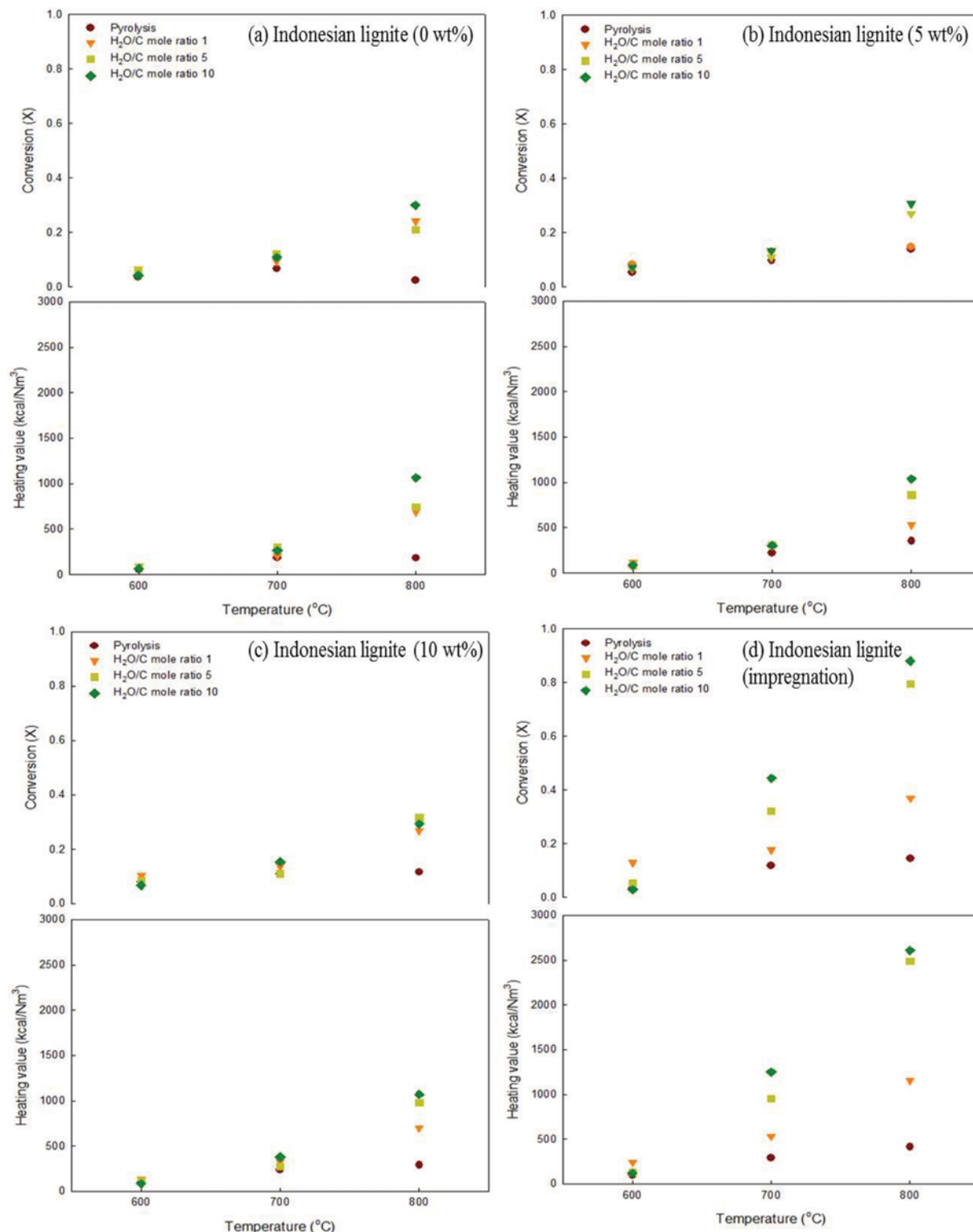


Fig. 5. Comparing effects of reaction temperature and catalyst addition on carbon conversion and heating value for Indonesian lignite.

for catalytic gasification.

Catalytic gasification with steam is a strong endothermic process because of the water gas and water gas shift reaction [30]. In a commercial plant (traditional coal to SNG plant efficiency is approximately 55%, GE Texaco, and Lurgi process coal gas efficiency

is 50-70%), the heating value and cold gas efficiency will be much lower than those of this study (the optimum condition cold gas efficiency is in the range of 70-80%) because the reaction will be formed using the heating value of coal itself rather than external heating [13,36,37].

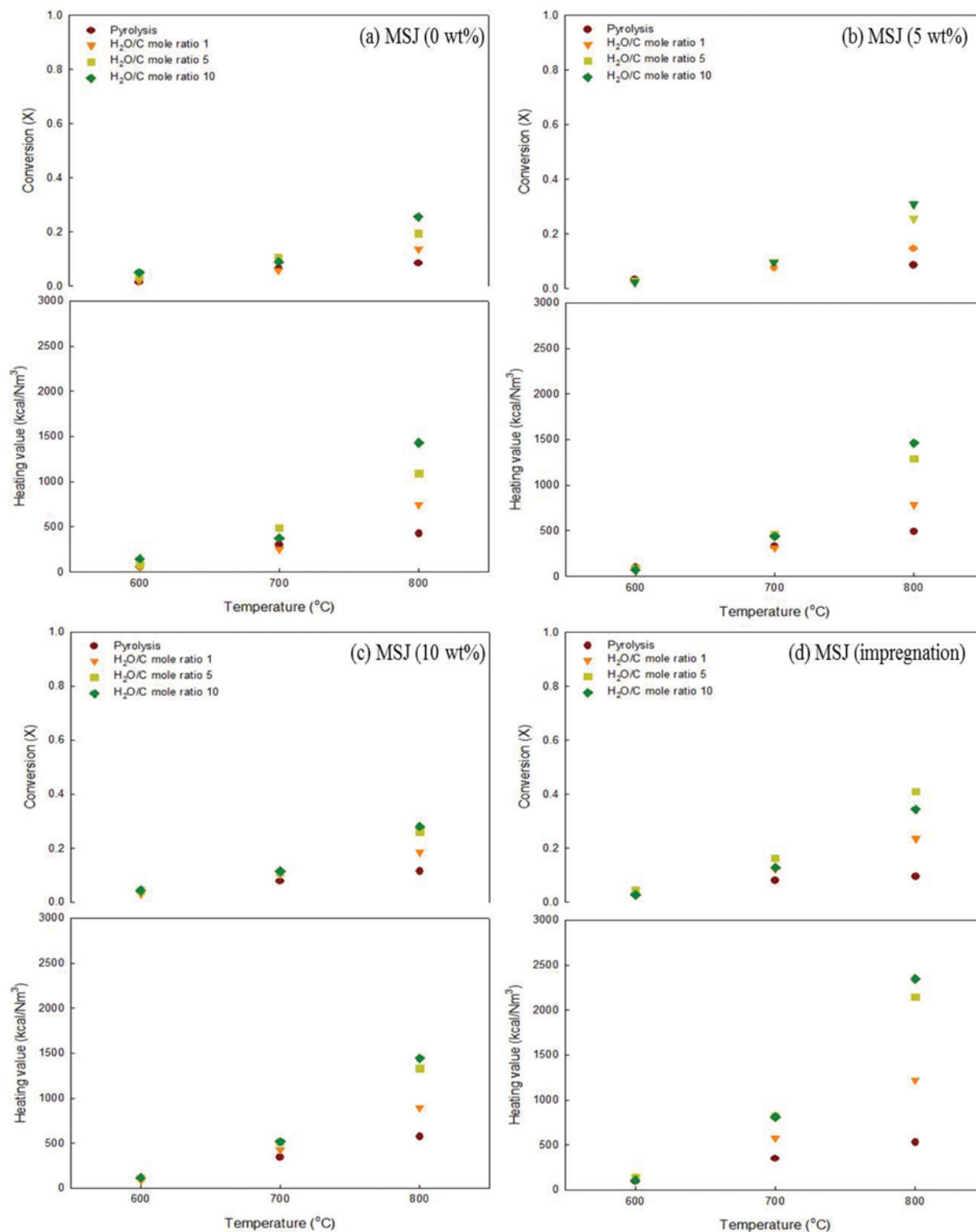
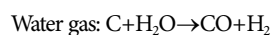


Fig. 6. Comparing effects of reaction temperature and catalyst addition on carbon conversion and heating value for MSJ.

3. Comparison Composition of Syngas Depends on the H₂O/C Mole Ratio

Fig. 8 shows that the composition of the syngas depends on the H₂O/C mole ratio. As the H₂O/C mole ratio increases, it is expected that the amounts of H₂ and CO will increase because of an improved water gas reaction; CO will decrease and CO₂ and H₂ will

increase because of the water gas shift reaction. In the non-catalytic or catalytic steam gasification, homogeneous and heterogeneous reactions produce syngas simultaneously as a major product with coal char, as shown in main reactions.



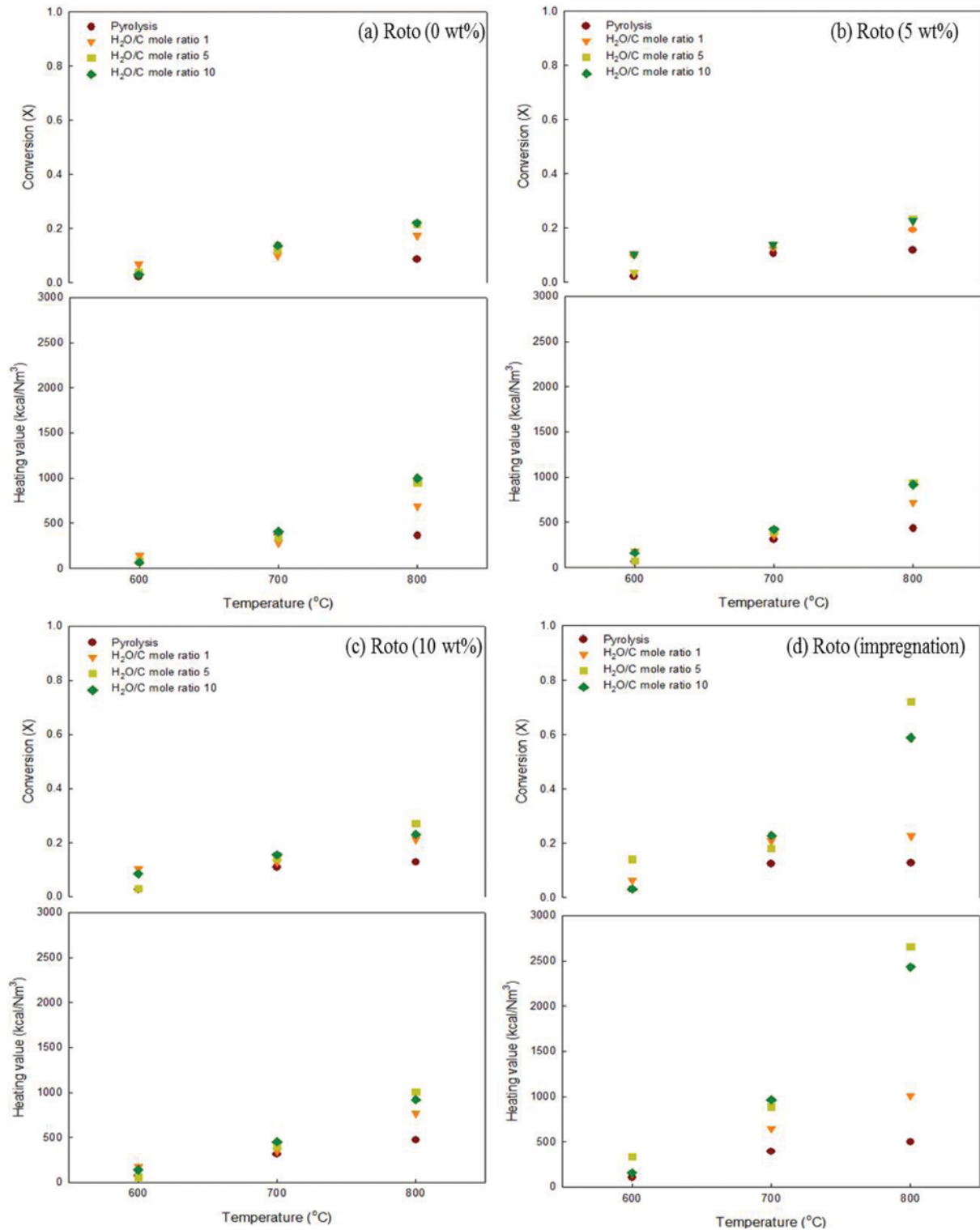


Fig. 7. Comparing effects of reaction temperature and catalyst addition on carbon conversion and heating value for Roto South.

Water-gas shift: $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$

As the $\text{H}_2\text{O}/\text{C}$ mole ratio increases, the amounts of H_2 and CO will increase because of an improved water gas reaction; CO will decrease and CO_2 and H_2 will increase because of the water gas shift reaction. In catalytic gasification, the main reaction may be

accelerated by active intermediate of metal catalyst. Also, this result shows no formation of CH_4 because of steam reforming reaction ($\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$) during the steam gasification. In this catalytic gasification, H_2 and CO_2 increased greatly and that of CO than non-catalytic gasification decreased greatly. From all the above results, this catalytic gasification could be considered to not only

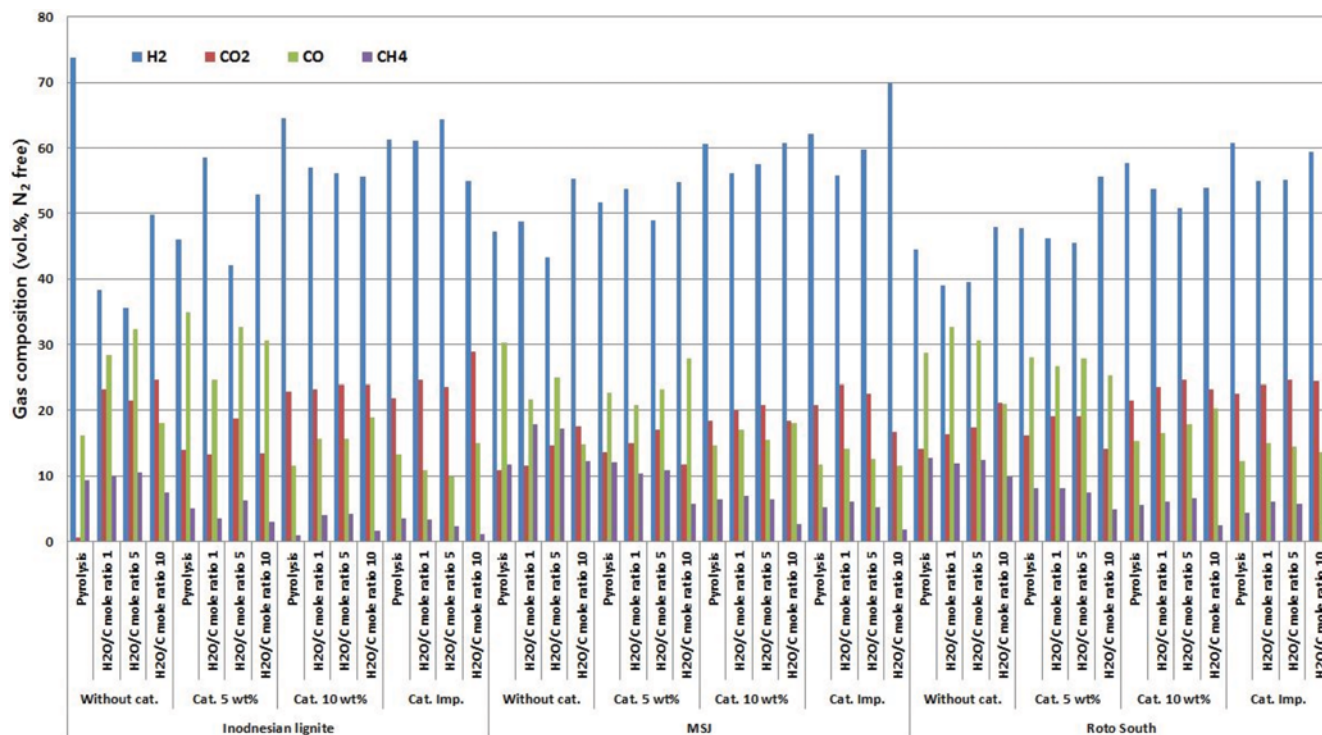


Fig. 8. Comparison of effects of H₂O/C mole ratio on gas composition with different catalyst addition in coal catalytic pyrolysis and gasification.

effectively accelerate the water-gas reaction, water-gas shift reaction, and steam reforming reaction of CH₄, but also to greatly enhance the produced H₂, with virtually no CH₄ formation [38].

This result is important because hydrogen evolution in the case of catalytic gasification is relaxed over a longer period of time compared to the hydrogen evolution from catalytic pyrolysis. This is due to the slower reaction kinetics of char gasification, which leads to the extension of the gasification process for a longer time duration [39]. At 800 °C, the results for both catalytic pyrolysis and gas-

ification (H₂O/C mole ratio: 10, impregnation coal) show that the hydrogen release in the case of catalytic pyrolysis is almost zero at 170 s, whereas the flow rate of hydrogen at 350 s in catalytic gasification with an H₂O/C mole ratio of 10.

As shown in Fig. 8, increasing the H₂O/C mole ratio causes increases in the H₂ composition and mole ratio of H₂/CO in the syngas [32]. This result is the same as that for syngas production using the catalytic steam gasification of municipal solid waste in a fixed-bed reactor [40]. Thus, the involvement of catalytic gasifica-

Table 5. Comparison of EDX data for char between catalytic pyrolysis and gasification at 800 °C

	H ₂ O/C mole ratio 0			H ₂ O/C mole ratio 1			H ₂ O/C mole ratio 5			H ₂ O/C mole ratio 10		
	Cat. 0 wt%	Cat. 10 wt%	Imp.	Cat. 0 wt%	Cat. 10 wt%	Imp.	Cat. 0 wt%	Cat. 10 wt%	Imp.	Cat. 0 wt%	Cat. 10 wt%	Imp.
Indonesian lignite												
C	99.04	97.81	11.91	98.45	98.71	37.06	100.00	99.38	22.22	99.75	91.99	71.98
K	0.96	2.19	45.37	1.55	1.29	27.00	-	0.62	43.41	0.25	8.01	28.02
O	-	-	42.72	-	-	35.94	-	-	34.37	-	-	-
MSJ												
C	77.09	79.43	37.22	79.79	71.41	17.76	82.09	83.51	17.10	82.24	81.38	62.04
K	-	4.74	29.35	0.55	5.37	42.48	2.06	0.50	37.28	1.30	1.27	14.95
O	22.91	15.83	33.43	19.66	23.22	39.76	15.85	15.99	45.62	16.45	17.34	23.01
Roto South												
C	100.00	37.61	47.76	97.92	99.08	36.64	77.16	99.58	38.97	99.17	98.71	26.12
K	-	2.39	52.24	2.08	0.92	32.41	1.96	0.42	27.39	0.83	1.29	36.24
O	-	-	-	-	-	30.94	20.88	-	33.63	-	-	37.64

tion in the water gas shift reversible reaction changes the composition so that H_2 is present in a higher amount than CO. Therefore, increasing the H_2O/C mole ratio led to direct increase in the H_2/CO ratio [41,42].

Table 5 show the results of an energy dispersive X-ray spectroscopy (EDX) comparison of the chars for catalytic pyrolysis and gasification at 800 °C. The overall EDX data provided in Table 5 verified the presence of only three major elements: carbon, oxygen, and potassium. The impregnated coal char contained potassium, but the other char has scarcely any potassium. Therefore, potassium was impregnated into the low rank coal of samples, and the K_2CO_3 played the role of a catalyst in the catalytic pyrolysis and gasification. The omnipresent coexistence of potassium, carbon, and oxygen indicated that the char was well-combined with the K_2CO_3 . This resulted in the consistent interaction of mineral matter with the K_2CO_3 in the catalytic gasification process, along with a loss in the catalytic activity and difficulty in recovering the catalyst [26].

CONCLUSIONS

We compared catalytic pyrolysis and gasification using the lab-scale bubble fluidized-bed reactor in order to utilize low rank coal, which is abundant and common around the world and whose price is low compared to bituminous coal as fuel. We analyzed the reaction characteristics of Indonesian three low rank coal as Indonesian lignite, MSJ, and Roto South to evaluate syngas composition by catalytic pyrolysis and gasification. The main results are as follows:

1) Catalytic gasification occurred by impregnating the catalyst in low rank coal, the catalytic gasification reactivity could be maximized even at a low temperature, and a high yield of hydrogen could be obtained.

2) Potassium was impregnated into the low rank coal, and the K_2CO_3 played the role of a catalyst in the catalytic pyrolysis and gasification.

3) Produced syngas tended to increase due to activation of reaction for all components as the temperature increased.

4) Because of the effect of catalyst, the syngas produced increased, but when the impregnation method was used instead of the physically mixing method, the H_2 produced was much better.

5) H_2O/C mole ratio and feed method of catalyst had significant effects on reactivity with respect to the catalytic pyrolysis/gasification.

6) The results of the lab-scale catalytic gasification showed that 70% of carbon conversion could be achieved with a shorter reaction within 6 min.

Future study will focus on increasing hydrogen yield, carbon conversion and decrease reaction time using mixed catalyst. Furthermore, the production of methane increased as the pressure increased based on the equilibrium reaction.

ACKNOWLEDGEMENTS

This work was supported by the New & Renewable Energy of the Korea Institute of Energy Technology Evaluation and Plan-

ning (KETEP) grant funded by the Korea government Ministry of Trade, Industry & Energy (No. 20113020050010).

This work was supported by Human Resources Program in Energy Technology of the Korea Institute of Energy Technology Evaluation and Planning (KETEP), granted financial resource from the Ministry of Trade, Industry & Energy, Republic of Korea (Project No: 2015 4010 200820).

REFERENCES

1. International Energy Agency, Energy Technology Perspectives (2014).
2. World Energy Resources: Coal World Energy Council (2013).
3. T. Takarada, Y. Tamai and A. Tomita, *Fuel*, **64**, 1438 (1985).
4. F. Huhn, J. Klein and H. Jüntgen, *Fuel*, **62**(2), 196 (1983).
5. N. C. Nahas, *Fuel*, **62**, 239 (1983).
6. T. Wigmans, R. Elfring and J. A. Moulijn, *Carbon*, **21**(1), 1 (1983).
7. D. W. McKee, C. L. Spiro, P. G. Kosky and E. J. Lamby, *Fuel*, **62**(2), 217 (1983).
8. K. J. Huttinger and R. Minges, *Fuel*, **64**(4), 486 (1985).
9. R. J. Lang, *Fuel*, **65**(10), 1324 (1986).
10. T. Takarada, S. Ichinose and K. Kato, *Fuel*, **71**(8), 883 (1992).
11. H. Kubiak, H. J. Schroter, A. Sulimma and K. H. van Heek, *Fuel*, **62**(2), 242 (1983).
12. L. Kuhn and H. Plogmann, *Fuel*, **62**(2), 205 (1983).
13. H. Juntgen, *Fuel*, **62**, 234 (1983).
14. K. Hashimoto, K. Miura and T. Ueda, *Fuel*, **65**(11), 1516 (1986).
15. S. J. Yuh and E. E. Wolf, *Fuel*, **62**(6), 738 (1983).
16. G. Bruno, M. Buroni, L. Carvani, G. Del Piero and G. Passoni, *Fuel*, **67**(1), 67 (1988).
17. A. Tomita, Y. Watanabe, T. Takarada, Y. Ohtsuka and Y. Tamai, *Fuel*, **64**(6), 795 (1985).
18. P. K. Bakkerud, *Catal. Today*, **106**(1), 30 (2005).
19. I. Handayani, A. Triantoro and D. Diniyati, *J. Novel Carbon Res. Sci.*, **7**, 68 (2013).
20. E. J Hippo and D. Tandon, *Preprints of Papers-american Chemical Society Division Fuel Chemistry*, **41**, 216 (1996).
21. H. Zhang, Dissertation at the Brigham Young University (2001).
22. S. Park, Y. Choi and J. Shon, *Appl. Chem. Eng.*, **22**(3), 312 (2011).
23. Y. T. Kim, D. K. Seo and J. H. Hwang, *Korean Chem. Eng. Res.*, **49**(3), 372 (2011).
24. D. W. McKee, *Carbon*, **20**(1), 59 (1982).
25. D. A. Sams, T. Talverdian and F. Shadman, *Fuel*, **64**(9), 1208 (1985).
26. J. Wang, K. Sakanishi and I. Saito, *Energy Fuels*, **19**, 2114 (2005).
27. L. Dong, C. Xu, T. Suda and T. Murakami, *Fuel Processing Technol.*, **91**(8), 882 (2010).
28. Fabrizio Scala, Woodhead publishing (2013).
29. W. J. Lee and S. D. Kim, *Fuel*, **74**(9), 1387 (1995).
30. J. Wang, M. Jiang, Y. Yao, Y. Zhang and J. Cao, *Fuel*, **88**(9), 1572 (2009).
31. O. C. Kural (Ed.), Istanbul Technical University, Istanbul (1994).
32. D. Tristantini, D. Supramono and R. K. Suwignjo, *Int. J. Technol.*, **6**, 22 (2015).
33. A. Kumar, D. D. Jones and M. A. Hanna, *Energies*, **2**(3), 556 (2009).
34. W. J. Lee, S. D. Kim and B. H. Song, *Korean J. Chem. Eng.*, **18**(5), 640 (2001).

35. J. M. Lee, Y. J. Kim and S. D. Kim, *Appl. Therm. Eng.*, **18**(11), 1013 (1998).
36. L. Chen, R. Nolan and S. Avadhany, MIT (2009).
37. S. Li, X. Ji, X. Zhang, L. Gao and H. Jin, *Appl. Energy*, **136**(31), 98 (2014).
38. Y. Wu, J. Wang, S. Wu, S. Huang and J. Gao, *Fuel Proc. Technol.*, **92**(3), 523 (2011).
39. I. Ahmed and A. K. Gupta, *Appl. Energy*, **86**, 1813 (2009).
40. S. Luo, Y. Zhou and C. Yi, *Energy*, **44**(1), 391 (2012).
41. L. Garcia, M. L. Salvador, J. Arauzo and R. Bilbao, *Energy Fuels*, **13**, 851 (1999).
42. J. G. Lee, J. H. Kim, T. J. Park and S. D. Kim, *Fuel*, **75**(9), 1035 (1996).