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Technical Report

Effect of K_2CO_3 as a Catalyst in Indonesian Low-Rank Coal Gasification on Product Composition

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The development of coal gasification has several disadvantages including high reaction temperature and big energy consumption, difficult purification of the gaseous products, and stringent requirements of the process equipment. It has also been pointed out to cause serious environmental pollution. To overcome these problems, the coal catalytic gasification technique can improve the reaction rate and conversion efficiency compared to conventional gasification techniques. In this study, the experiment was conducted by flowing nitrogen, oxygen, and steam, into the gasification equipment. The gasification reactor was heated to 600 °C and 800 °C, where dry coal had been initially fed into before the gasification process began. The flow rates of pyrolysis products were then measured and a sample was taken for further analysis by using a gas chromatograph. The analysis identified the products composition that was CO, CO_2, H_2, CH_4, C_2H_4 and C_2H_6. The experiments that used K_2CO_3 as a catalyst were conducted by feeding the catalyst into the reactor. After the catalyst started to react, the coal was slowly fed. The composition of synthesis gas was observed to depend on the gasification agent used. Based on the experiment of synthesis gas production from Indonesian low-rank coal with catalyst K_2CO_3, it can be summarized that there is a good correlation between gasification agent, coal and catalyst used, with the quality and quantity of synthesis gas produced.

1. Background

Low-rank coal is mostly not yet exploited due to the high moisture content and low calorific value. Thus, this non exportable coal is potential for local utilization to produce electricity, to be upgraded into good quality coal, or to be converted into other valuable products. To this end, intensive research and development programs have to be carried out with reasonable objectives. One of these could be to secure future energy and raw material supplies for industries in attempt to preserve the oil reserves[1)].

The gas production from coal, known as town gas, was practiced in Indonesia until early 1970's by using special imported high-rank coal, *i.e.* coking coal. Then, gasification of Indonesian non coking coal by utilizing fixed bed technology has started since 1995 for the generation of producer gas[2)].

The properties of gases produced from coal gasification depend on both types of coal and gasification agents. Coal gasification can be defined as the conversion of coal into gaseous products, especially carbon monoxide (CO) and hydrogen (H_2), with or without the use of a reactant or a gasification agent (air, oxygen, steam, CO, or mixture of them), in a reactor. If the gasification is carried out directly in-situ, such as in a coal seam, the process is known as underground coal gasification. CO and H_2 can then be processed into methane that is known as synthetic natural gas (SNG). The methane yielded from the gasification process is different from coal bed methane (CBM) since CBM is a trapped methane which already presences in the coal seam during the coalification process[3)].

In line with a governmental program to develop clean coal technology and to apply more stringent environmental regulations, this conversion of coal into gaseous products could be alternative utilization of Indonesian low-rank coal. The gas obtained from coal gasification can be used as fuel and as raw material for chemical industries as well as for synthetic oil.

Pertaining to the objective of securing energy and raw material supplies for industries, a research on coal gasification of Indonesian low-rank coal for generating synthesis gas is required. Thus, in this study, the experiment was conducted by employing Indonesian low-rank coal, catalyst K_2CO_3, and a fluidized bed reactor.

Nevertheless, the development of coal gasification has several disadvantages, including high reaction temperatures and big energy consumption, difficult purification of the products, stringent requirements of the process equipment. Additionally, it has also been pointed out to cause serious environmental pollution. To resolve some of these issues, coal catalytic gasification technique has been developed to improve the reaction rate and conversion efficiency compared to conventional gasification techniques[4)].

2. Coal and catalyst used

A sample of low-rank coal from Aceh, Sumatra, was used for this experiment. Initially, it was crushed into 20 mesh size and was then dried at 100 °C prior to being fed into the reactor. The properties for this coal sample are shown in Table 1.

Table 1 The properties of sample coal

Analytical Parameter	Result	Unit	Basis	Reference Standards
Moisture in Air Dried Sample	9.74	%	adb	ASTM D. 3173
Ash	14.38	%	adb	ASTM D. 3173
Volatile Matter	40.58	%	adb	ISO 562
Fixed Carbon	35.30	%	adb	ASTM D. 3173
Calorific Value	4890	Cal/g	adb	ASTM D. 5865
Total Sulfur	0.14	%	adb	ASTM D. 4239
Carbon	56.10	%	adb	ASTM D. 5373
Hydrogen	5.19	%	adb	ASTM D. 5373
Nitrogen	1.16	%	adb	ASTM D. 5373
Oxygen	23.03	%	adb	ASTM D. 3176

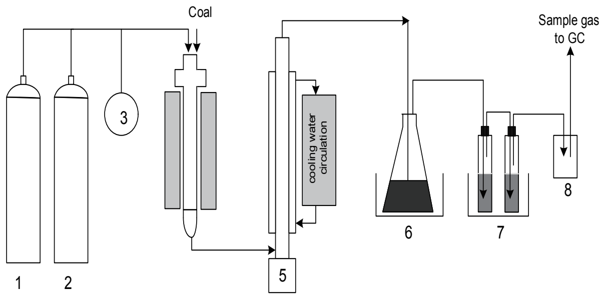
3. Experimental methods

Coal gasification process without catalysts was conducted by using silica sand particles as bed materials, which were initially charged into the reactor. It was then followed by the injection of nitrogen. The reactor was preheated to approximately 600 °C and 800 °C, desirable temperatures for taking gas data. Subsequently, coal and gasification agent were then fed into the bed. The flow rate of reactant used for gasification was calculated based on the stoichiometric coefficients. Besides coal, other variables used in the experiment were reactants that included oxygen, steam, and mixture of oxygen and steam.

Experiments using K_2CO_3 catalyst were conducted by feeding the catalyst into the reactor with mass ratio 5% of total mass of coal. After the catalyst started to react, the coal was slowly fed into the reactor, with the total mass of the coal was 20 grams. The time required for one cycle of experiment was 2-3 hours.

Gasification reactor made of quartz glass is equipped with an electric heater. Heat exchanger serves to cool the products of volatile matter. Bottle condensation serves to catch water and tar in the gas. Pockets of gas sampling and gas chromatography used for gas composition analysis.

The experiment used oxygen as the gasification agent. Both oxygen and nitrogen were flowed to a series of experimental tools. Nitrogen flow rate was set at 200 mL/min while oxygen (O_2) was at 50 mL/min. In the experiment that used the mixture of steam and oxygen as gasification agent, nitrogen flow rate was set at 150 mL/min, oxygen was at 50 ml/min and 50 mL/min steam. For steam as sole gasification agent, nitrogen flow rate was set at 200 mL/min and steam was at 50 mL/min. Note that, gasification reactor was heated to 600 °C and 800 °C while heat exchanger (HE), condensation bottles (containing 60 mL of distilled water) and tar wash bottles (containing 60 mL anisole) were cooled to 10 °C and -4 °C, respectively. Dry coal was fed into the reactor prior to the gasification process. Pyrolysis gasification process was carried out for 10 minutes. The products were then sampled by a gas sample bag and were then analyzed using gas chromatography. The compositions results were CO, CO_2, H_2, CH_4, C_2H_4 and C_2H_6. In the end of experiment, the pyrolysis reactor was cooled to 40 °C and the charcoal formed was then weighed.



1. Oxygen tube 2. Nitrogen Tube
3. Steam Reactor 4. Reactor
6. Condensation bottle 5. Heat exchanger
7. Tar wash bottles 8. Gas sample bag

Fig. 1 The scheme of Coal Gasification Equipment Employed.

4. Result and discussion

4.1 Effect of the gasification agents on the gasification products

The compositions of synthesis gas yielded from different gasification agents without catalyst are depicted in Figs. 2 and 3. It can be seen that the product compositions depend on the gasification agent used. The gasification without catalyst at 600 °C using oxygen as gasification agent produced synthesis gas with the highest CO content, up to 27.78%, with low content of H_2 (3.41%). The gasification using oxygen/steam mixture could generate gas with the highest CO content of 19.33%, with low content of H_2 (4.81%). Meanwhile, steams blown gasification produced the highest H_2 content of 28.12% and low CO content (9.07%).

Gasification without catalyst at 800 °C using oxygen as gasification agent produced a synthesis gas with the highest H_2 content of 9.75%, but with low content CO content (6.56%). Gasification using oxygen/steam mixture produced gas with highest H_2 content of 44.97% but with low CO content (9.34%). Meanwhile, steams blown gasification produced the highest H_2 content of 46.04% with low H content (10.85%).

The compositions of synthesis gas resulted from different gasification agents with catalyst are presented in Figs. 4 and 5. Similar to gasification without catalyst, the composition of synthesis gas depend on the gasification agent used.

Gasification with a catalyst K_2CO_3 at 600 °C using oxygen as gasification agent produced synthesis gas with the highest CO content of 33.09%, but with low H_2 content (5.80%). Gasification using oxygen/steam mixture yielded gas with the highest CO content of 30.62%, but with low H_2 content of 13.57%. Meanwhile, steams blown gasification could generate the highest H_2 content of 22.74% and low CO content of 11.63%.

Gasification with catalyst K_2CO_3 at 800 °C using oxygen as gasification agent produced synthesis gas with the highest CO content of 16.93%, with low H_2 content of 9.04%.

Gasification using oxygen/steam mixture could produce gas with highest H_2 content of 19.60%, but with low CO content of 11.03%. Meanwhile, steams blown gasification produced the highest H_2 content of,28.58% and low H_2,content of 14.86%.

Principally, oxygen blown gasification is a partial combustion of coal in which carbon reacts with oxygen to form CO, while in steam blown gasification there is a shift reaction between steam and hydrocarbons and also char from coal to form most hydrogen. The reactions of oxygen blown and steam blown gasification are as follows[5] :

$C + ½ O_2 \longrightarrow CO$ (partial combustion)

$C + H_2O \longrightarrow CO + H_2$ (shift reaction)

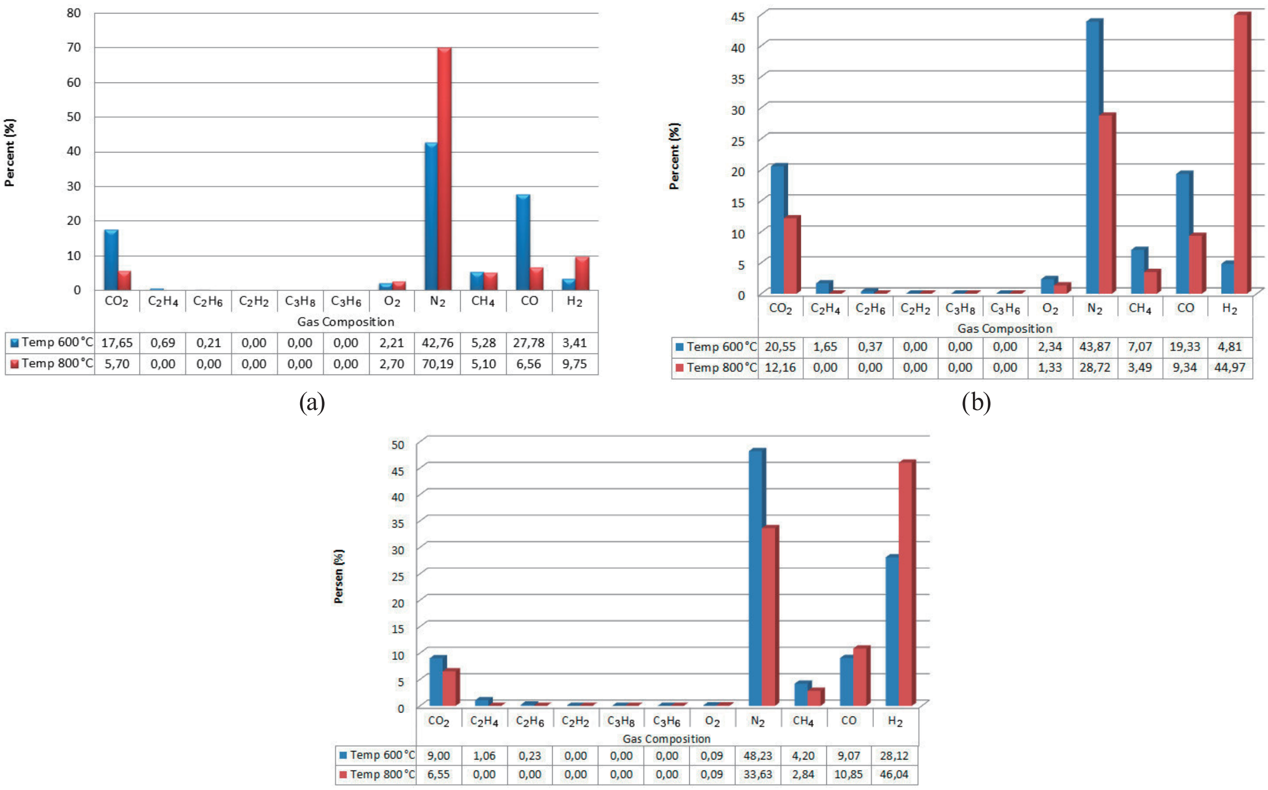


Fig. 2 (a) Gasification with oxygen as gasification agent without catalyst,
(b) Gasification with oxygen and steam as gasification agent without catalyst,
(c) Gasification with steam as gasification agent without catalyst.

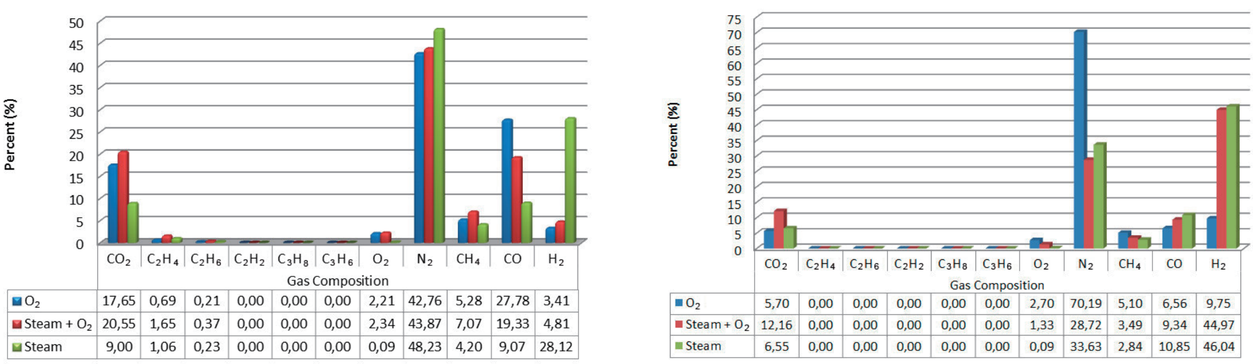


Fig. 3 (a) The effect of gasification agent on gas composition without catalyst at 600 °C,
(b) The effect of gasification agent on gas composition without catalyst at 800 °C.

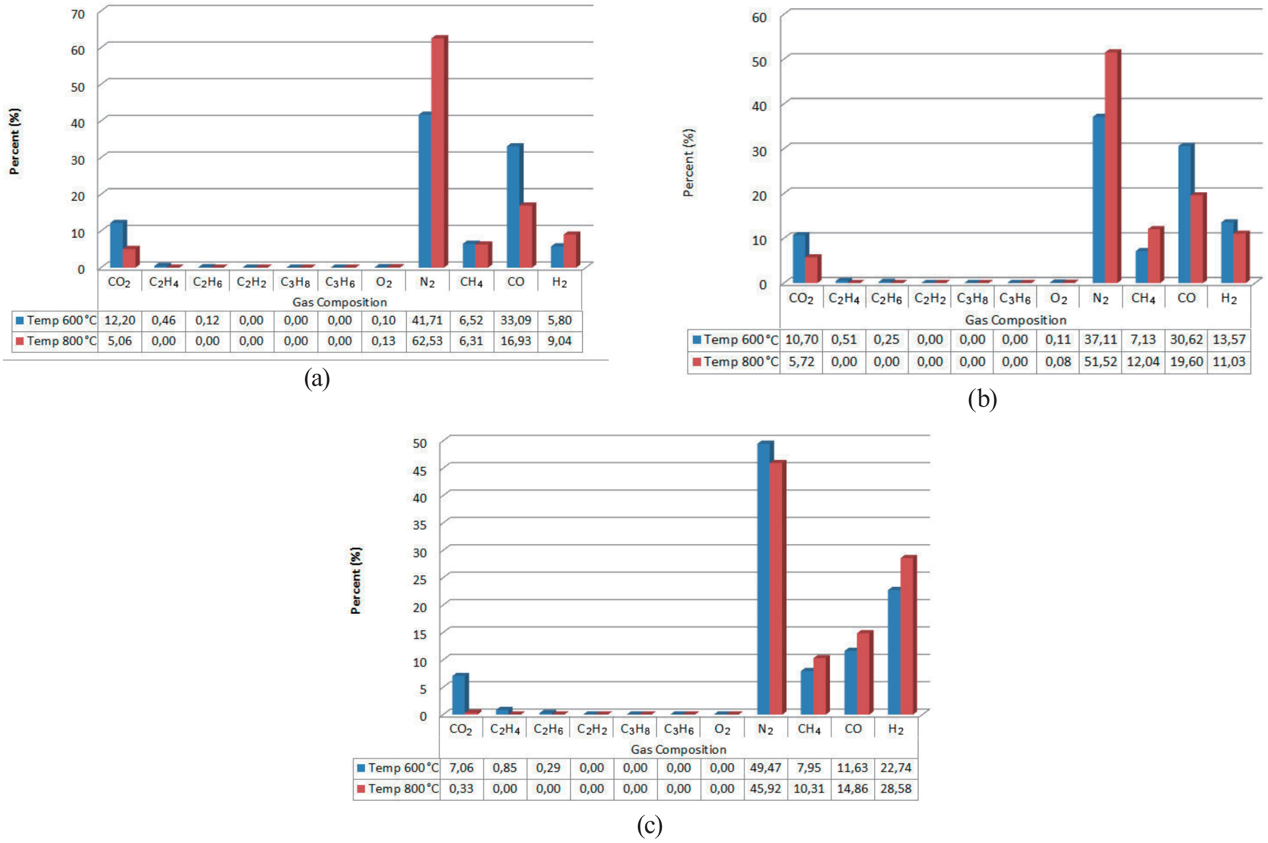


Fig. 4 (a) Gasification using oxygen as gasification agent with catalyst K_2CO_3,
(b) Gasification using oxygen and steam as gasification agent with catalyst K_2CO_3,
(c) Gasification using steam as gasification agent with catalyst K_2CO_3.

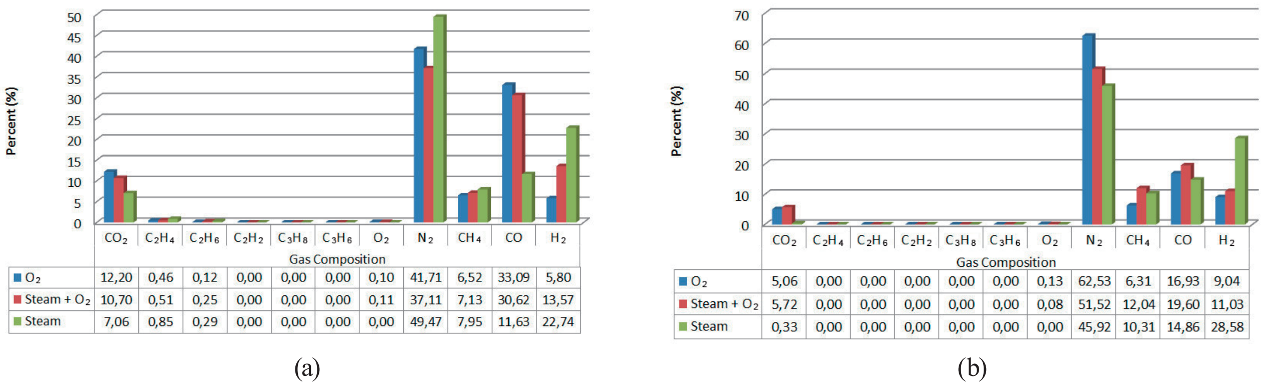


Fig. 5 (a) The effect of gasification agent on gas composition with catalyst K_2CO_3 at 600 °C,
(b) The effect of gasification agent on gas composition with catalyst K_2CO_3 at 800 °C.

4.2 Effect of gasification agents on the net heating value of gas produced

Fig. 6 shows the effect of gasification agents on net heating value (NHV) of gas produced without catalyst K_2CO_3. Oxygen blown gasification at 600 °C produced gas with the highest NHV, *i.e.* 5,773.61 kJ/m^3 while steam/oxygen mixture blown gasification produced gas with the lowest NHV *i.e.* 5,492.49 kJ/m^3. Meanwhile, gasification using steam yielded NHV value of 5,672.76 kJ/m^3, which was between the highest and lowest NHV values observed. It indicates that gas produced from oxygen blown gasification containing higher CO since the CO has higher NHV than H_2.

Meanwhile, at 800 °C with steam as gasification agent, the highest NHV obtained was 7,340.16 kJ/m^3 and oxygen blow gasification produced gas with the lowest NHV of 3,704.53 kJ/m^3. It implies that the gas produced at the temperature of 600 °C has smaller NHV than the one produced at 800 °C because gas from gasification contains higher H_2 that has lower NHV than CO.

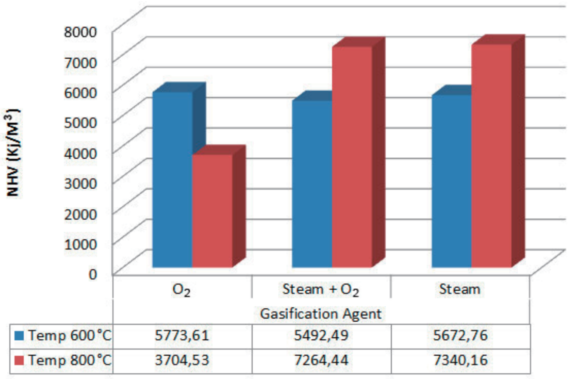


Fig. 6 The NHV of gases produced by gasification using different gasification agent without catalyst.

Fig. 7 shows the effect of gasification agents on net heating value (NHV) of gas produced with catalyst K_2CO_3. It was observed that oxygen/steam mixture blown gasification at 600 °C produced gas with the highest NHV of 7,884.45 kJ/m^3 and steam blown gasification produced gas with the lowest NHV of 6,762.45 kJ/m^3, while gasification using oxygen produced gas with NHV value of 7,143.98 kJ/m^3, which is within the range of the highest and lowest values. It suggests that the use of catalyst K_2CO_3 catalysts could increase the net heating value (NHV).

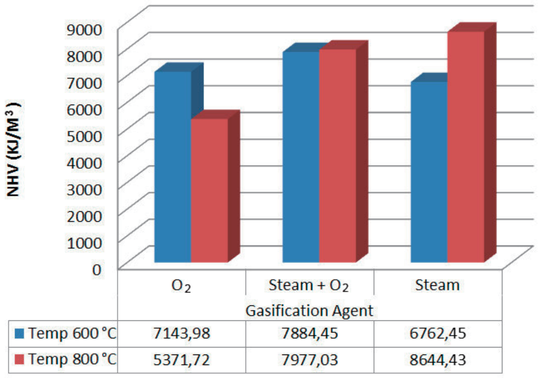


Fig. 7 The NHV of gases produced by gasification using different gasification agents with catalyst K_2CO_3.

4.3 Effect of gasification agents on the cold gas efficiency and carbon conversion

Cold gas efficiency could be defined as the ratio of the energy content of producer gas and the energy content of feed coal[6)]. Fig. 8 shows the effect of different gasification agents on the cold gas efficiency without catalyst K_2CO_3.

The highest efficiency of cold gas without catalyst could be obtained at the temperature of 800 °C with steam gasification agent (33.24%) while oxygen blown gasification produced gas with the lowest cold gas efficiency (16.78%) at 800 °C. This could be explained by the higher NHV value with steam as gasification agent compared to oxygen at 800 °C.

Fig. 9 shows the effect of different gasification agents on the cold gas efficiency with catalyst K_2CO_3.

The highest efficiency of cold gas with catalyst was obtained at the temperature of 800 °C with steam as gasification agent (39.15%) while oxygen blown gasification could only produce gas with the cold gas efficiency of 24.33%. at 800 °C.

Fig. 10 shows the effect of different gasification agent on carbon conversion for gasification without catalyst K_2CO_3. The highest carbon conversion (55.21%) was observed at the temperature of 600 °C with oxygen as gasification agent while the lowest (18.90%) was obtained with similar gasification agent at 800 °C.

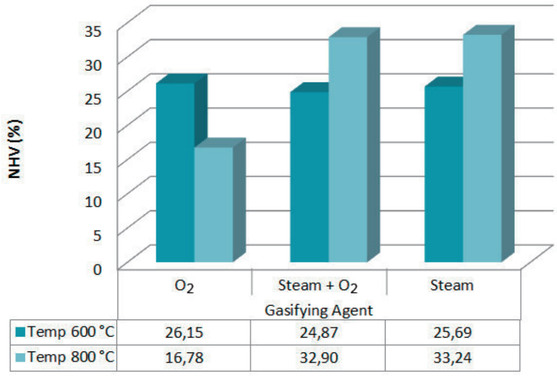


Fig. 8 Cold gas efficiency without catalyst K_2CO_3.

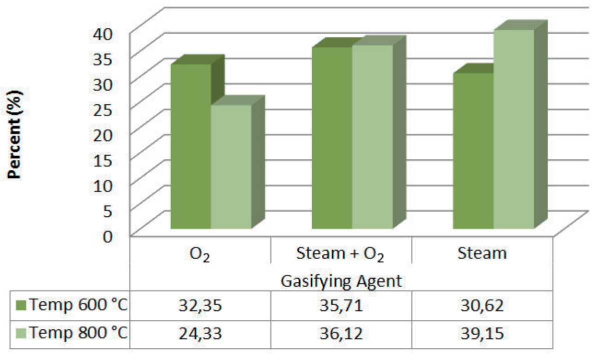


Fig. 9 Cold gas efficiency with catalyst K_2CO_3.

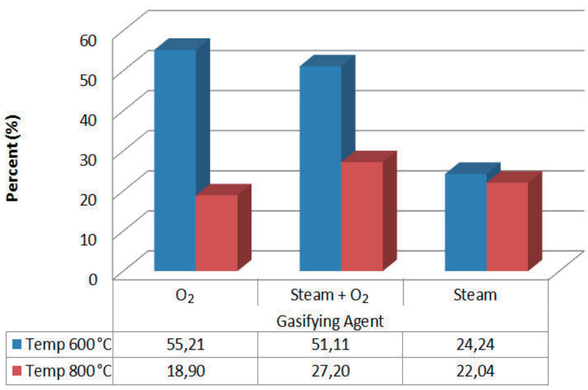


Fig. 10 Carbon conversion without catalyst K_2CO_3.

Fig. 11 shows the effect of different gasification agent on carbon conversion with catalyst K_2CO_3. The highest carbon conversion (56.41%) was obtained at the temperature of 600 °C with oxygen as gasification agent while the lowest (27,76%) was obtained at 800 °C with steam as gasification agent.

Thus, it could be summarized that the addition of K_2CO_3 catalyst to the coal gasification process could increase the efficiency of gasification.

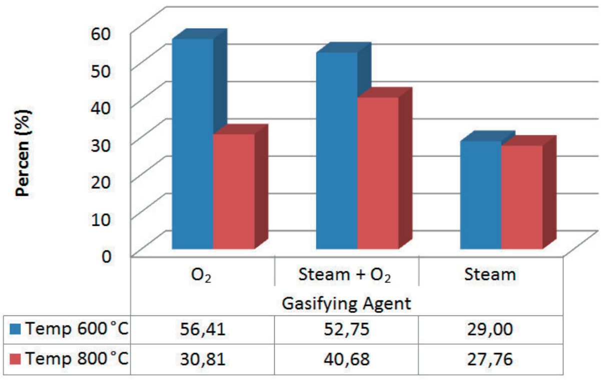


Fig. 11 Carbon conversion with catalyst K_2CO_3.

5. Conclusion

A study on the production of synthesis gas from Indonesian low-rank coal using catalyst K_2CO_3 has been conducted. The results show that there is a good correlation between gasification agent, coal and catalyst used, with the quality and quantity of synthesis gas produced.

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