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The Effect of Temperature and Time of Gasification Process and The Addition of Catalyst to The Composition of The Combustible Gas from The Wastes of Tobacco Leaves With Gasifier Updraft

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INFORMASI ARTIKEL

ABSTRAK

The increasing industrial production is causing more of energy consumption needed. Tobacco leaves are waste from cigarette production that can be use as alternative biomass energy to solve the problems of the energy crisis in the near future. Gasification is the conversion of solid carbon raw materials from biomass by the partial oxidation to be combustible gas (CO, H_2, and CH_4) as an energy source. This research uses the fixed-bed reactor with capacity of 300 grams by the flow of updraft and raw material of waste tobacco leaves that have a size -14 + 18 mesh. The influence of temperature was research on 700 °C, 800 °C, and 900 °C with the reaction time for 20 minutes and the addition of a catalyst K_2CO_3. gasification medium used air and steam with the constant flow rate 0.504 m3/hour air and 0.0555 kg/minutes steam. Step of the process with the preparation of raw materials, the gasification process, and analysis of gas products. The result of the gasification process analysed using the gas chromatography, and the best results were obtained on the temperature 700 °C in the process time for 10 minutes produced combustible gas are CO = 22.82 % v, H_2 = 26.82 % v, CH_4 = 7.09 % v. increasing production gas happened when a catalyst K_2CO_3 is added, the producting of combustible gasses are CO = 52.76 % v, H_2 = 23.46 % v, CH_4, = 3.23 % v

Keyword: waste tobacco leaves , gasification, catalyst , updraft gasifier , combustible gas.

1. INTODUCTION

Industrial production in Indonesia is increasing every year so causing more energy consumption. Based on data from the Directorate General of Renewable Energy and Energy Conservation of the Ministry of Energy and Mineral Resources, in recent year Indonesia's energy consumption growth reached 7% per year [KemenESDM, 2018]. Reliance on conventional fuel, especially in Indonesia, has reached an alarming level and one of the ways to solve the energy crisis by utilizing renewable alternative energy sources.

Java Island is the largest producer of tobacco with 135.286 hectares of land, and production of dried leave 114.528 tons in 2013 and predicted in 2015 will reach 116.582 tons production [Kemenprint, 2012]. If it is considered 26% of total weight of the tobacco leaves are the stem and leave stalk, so the production waste will be 30,311 tonnes per year.

The tobacco leave stalk is a waste of cigarette production that can be used as an alternative to solve the energy crisis problem in the near future because it's very abundant. The needing of cigarette consumption from years to years is increasing. Based on Kementerian Perindustrian's data, cigarette production has grown in the range of 5% - 7.4% per year. In 2015, cigarette production is predicted will reach 398.6 billion

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cigarettes, and In 2016 is expected it will rise 5.7% - 421.1 billion cigarettes [Kemenprin, 2015].

The waste is only used as compost and fuel in the direct combustion process or take nicotine content to produce pesticide now. Therefore, special technology is needed to increase the economic value of wastes tobacco leave become renewable alternative energy sources. In this research, the researcher used gasification method because it's more 8% saving cost and also 12% saving fuels than direct burn [Jayah T.H, 2003]. Gasification is a process for converting solids become combustible gases (CO, CH_4, and H_2) by combustion processes in high temperatures on gasifier reactor with limited air supply, the gas process can be used as fuels, chemicals or to producing electrical energy.

2. THEORETICAL BASIC
2.1 Gasification

Gasification is a thermochemical process that converting carbon material such as biomass become useful gas fuel or chemical raw material [Basu, 2010]. The gasification process generally consists of the drying process, pyrolysis, oxidation, and reduction can be seen in Figure 1.
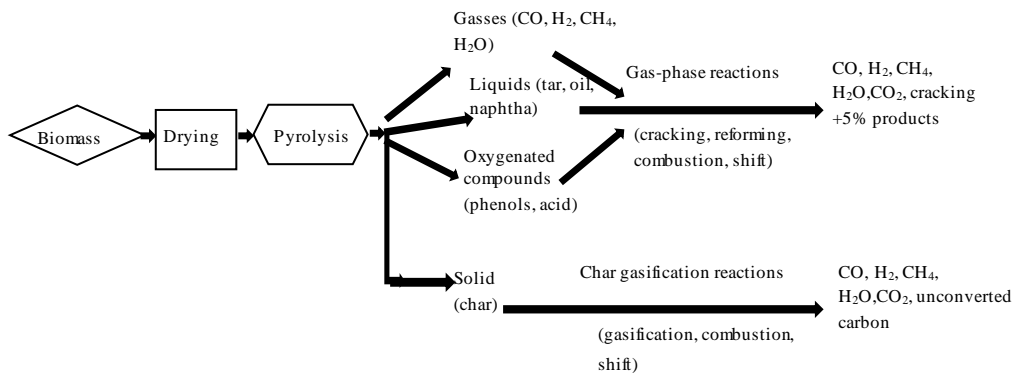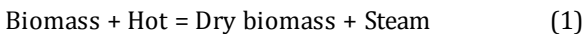

Fig. 1. Gasification process

The Stage of Gasification

1. Drying Zone

The drying takes place at a temperature of 100°C and it will continue until a temperature of 250°C is reached [13]. This process will vaporize most of the water content in the feedstock. In this zone, there is no chemical change except water evaporation.

$$\text{Biomass} + \text{Hot} = \text{Dry biomass} + \text{Steam} \qquad (1)$$

2. Pyrolysis zone

Pyrolysis or devolatilization can be referred to partial gasification. A chain of physical and chemical processes occurs during the pyrolysis process. The pyrolysis process begins at a temperature of 250-500 °C. when thermally components unstable, such as lignin in biomass rupture and evaporate in conjunction with other components. Evaporating of liquid products contains tar and PAH (polyaromatic hydrocarbon). Generally, pyrolysis products consist of three types, namely gas, tar, and charcoal. There is a reaction generally, that occur in pyrolysis, and the products are:

$$\text{Biomass} \rightarrow \text{Charcoal} + \text{Water vapor} + \text{Gas} + \text{Tar} \quad (2)$$

3. Zone of oxidation or combustion (Combustion)

The gas of the pyrolysis stage will then be oxidized by oxygen from the gasification medium. The temperature in the lower layers far exceeds the carbon ignition temperature, so the combustion reaction is very exothermic, due to the excess oxygen. The heat released heats the moving gas up and solids down. The reaction is [Susanto, 2004]:

$$C + O_2 \rightarrow CO_2 \ (\Delta H = - 394 \text{ kJ / mol}) \qquad (3)$$
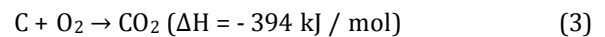
The combustion reaction (equation 3) takes place very quickly, this reaction consumes most of the available oxygen. Since the available oxygen is reduced, the combustion reaction turns to partial combustion, releasing CO and a moderate amount of heat [Basu, 2010]. Here's the reaction:

$$C + \tfrac{1}{2} O_2 \rightarrow CO \ (\Delta H = - 111 \text{ kJ / mol}) \qquad (4)$$
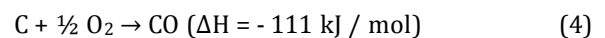
4. Reduction Zone

The hot gas, a mixture of CO, CO_2, and steam from the feed and gasification media, moves into the gasification zone (reduction), char from the gasifier bed is classified with CO2 entering the reduction zone, resulting in a decrease in CO2 concentration in the gasification zone [Susanto, 2004].

$$C + CO_2 \rightarrow 2CO \ (\Delta H = +172 \text{ kJ / mol}) \qquad (5)$$
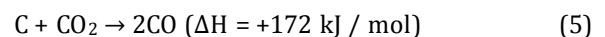
The producer gas reaction is an endothermic gasification reaction, which produces hydrogen and carbon monoxide from carbon. The gas mixture of this product is also known as synthesis gas, or syngas:

$$C + H_2O \rightarrow CO + H_2 \ (\Delta H = +131 \text{ kJ / mol}) \qquad (6)$$

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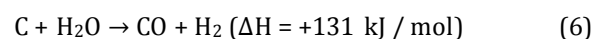
Gasification process stages that take place on the gasifier can be observed in the temperature range of each zone :
1. Drying: T> 100 ^{o}C
2. Pyrolysis: 250 ^{o}C <T = 500 ^{o}C
3. Oxidation: T = 1200 ^{o}C
4. Reduction: 600 ^{o}C <T <1000 ^{o}C

2.2 Factors affecting the gasification process
1. Time

The longer the gasification time, the more gas produced. However, this depends on the fuel mass used. In a downdraft gasifier, the effective time of gasification of waste of palm starch with a weight of 3 kg of material is about 1.5 hours to produce gas.

2. The Energy content of raw materials used

Raw materials with high energy content will provide better gas combustion. The higher the energy content of the raw materials, the higher the syngas gasification results because the energy that can be converted is also higher.

3. The Water content of raw materials

The higher the water content of a material, the more heat it will take to evaporate water. Pyrolysis products consist of solids, water, and liquid products with the greatest percentage of water. This is due to the high water content of the bait. Raw materials with lower moisture levels will be easier to classify than raw materials with high moisture levels.

4. Temperature is very influential on the reaction speed.

The temperature gasification depends on the combustion speed of the raw material and the rate of air. In steam gasification, the chemical reaction runs optimally at a temperature range of 900-1200 ^{o}C. The temperature of the reactor when the gasification process takes place greatly affects the production of the resulting gas. For that, the gasification reactor needs to be insulated to maintain the temperature inside the reactor remains high.

5. The size of the raw material

The effect of porosity, the size of the raw material plays an important role in the distribution of heat in the reactor, ensures the continuity of the reaction quickly and effectively, separation of solids and the collection of good liquid products [Diebold, 1999]. The size of the feedstock also affects the speed of mass reduction and loss of fluids and or gases, besides the smaller raw material size requires a fan or blower with higher pressure. Raw materials of too small size will result in decreased pressure and the amount of impurities in the outflow gas resulting in low gasification temperature resulting in a lot of tar due to the decrease in pressure plays a role in reducing the gas load in the gasifier equipment while the raw material of too large size will reduce fuel reactivity because it creates problems on

startup and a little gas condition. Each type of gasifier has a range of different feedstock sizes, for fixed bed gasification particle diameters between 20 mm - 100 mm and length of 1/10 - 1/5 of bed diameter [Kaup, 1981].

6. Airflow velocity

On the gasification, the increase in airflow speed will accelerate the combustion process and reduce the yield of solids. The greater the air rate the fuel consumption rate will be greater and the process time will be shorter. The addition of large amounts of oxygen into the reactor causes the material to burn faster to charcoal. In the process of downdraft gasification of tobacco leaf waste, the highest syngas concentration was found in variation Q = 3 m^3/h at minute 30 with CH_4 syngas concentration of 2.27% vol, CO gas 7.17% vol and H_2 gas 5 , 79% vol [Suhendi, *et al*, 2016] .

7. The Ratio of air and raw materials (AFR)

Comparison of air and raw materials in the process of gasification affect the reaction that occurs and of course on the resulting syngas content. Air requirements in the gasification process vary, according to the energy content present in the raw materials. Therefore a precise ratio is required if a maximum syngas yield is desired.

2.3 Catalyst
The catalyst is defined as a substance that can accelerate the reaction rate toward the equilibrium, without the catalyst being consumed in the process. The use of catalysts in the thermochemical conversion of biomass may not be important, but it can help in certain circumstances. The two main motivations for the catalyst are [Basu, 2010]: 1. Removal of the tar from the product gas, especially if the downstream application or installed equipment can not tolerate it. 2. The reduction of the methane content of the product gas, especially if it is used as a syngas (a mixture of CO, H_2). The development of catalytic gasification is driven by the need for reform tar, when product gas passes through catalyst particles, tar or hydrocarbons can be condensed Alkali metal catalysts Potassium carbonate and sodium carbonate the most widely used in biomass gasification as the main catalyst. K_2CO_3 is more effective than Na_2CO_3. Unlike dolomite, they can reduce methane in gas products through reform reactions. Many types of biomass have potassium inherent in their ashes, so they can benefit from potassium catalytic action with reduced tar production

3. MATERIALS AND METHODS

3.1 Preparation Of Raw Material
In this process the tobacco leaf waste is soaked with water to remove the nicotine content contained in the material, then the waste of tobacco leaves is dried with sunlight to reduce the water content, then the tobacco leaf waste is inserted into the crusher. After that screening to get the desired material size.

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3.2 Gasification Process

Before the gasification process begins, first prepare gasification tools, teh gasification begins by loading the raw material into the reactor of 300 gram. Then turn on the compressor and adjust air flow rate by the flow meter and obtained the air flow rate of 0,504 m3/hour. Turn on the heater and set temperature that has been determined. make steam using water, and open when it reaches the point of temperature. and turn on the condenser.

During gasification process record and observation temperature process every minutes. if the temperature has reached its point then take the gas with sample bag every five minutes. After gasification complete turn off heater, compressor, and condenser, and take the tar sample from bottom condenser with the bottle. After that, sample gas analyzed using Gas Chromotography. The scheme of gasification tools can be seen in Fig. 2.
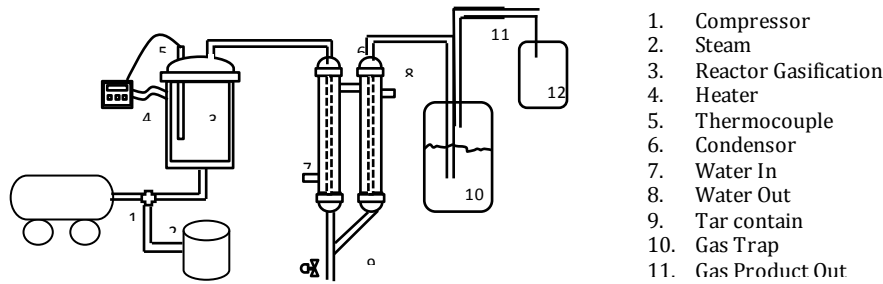
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1. Compressor
2. Steam
3. Reactor Gasification
4. Heater
5. Thermocouple
6. Condensor
7. Water In
8. Water Out
9. Tar contain
10. Gas Trap
11. Gas Product Out

Fig. 2. Scheme Gasification Tools

4. RESULTS

Biomass components are analyzed first, using the ultimate analysis performed at PT. Indonesia Power and proximate analysis conducted at TekMIRA Laboratory. The results of biomass analysis can be seen in Tabel 1.

Tabel 1. Results of ultimate analysis and proximate of waste tobacco leaves

Ultimate Analysis	Results (wt%)*	Proximate Analysis	Results (wt%)
Karbon (C)	36.85	Moisture	12.98
Hidrogen (H)	3.85	Ash Content	19.83
Sulfur (S)	0.12	Volatile Matter	55.09
Oksigen (O)	28.31	Fixed Carbon	12.10
Nitrogen (N)	0.54		

*Suhendi, et, al. 2016

4.1 Temperature Profile

Gasification reactors have four zones: drying zone, pyrolysis, oxidation, and reduction. Each stage or zone has a range of different temperatures, the temperature achievement of the gasification process affects the resulting fuel yield. The result of the temperature achievement process can show the zones present in the gasification. Fig. 3. shows the temperature profile contained in the gasification reactor. indicates that the process is in the drying zone at 60 - 2600 seconds, wherein the content of H_2O owned by tobacco leaf waste is removed by evaporation.
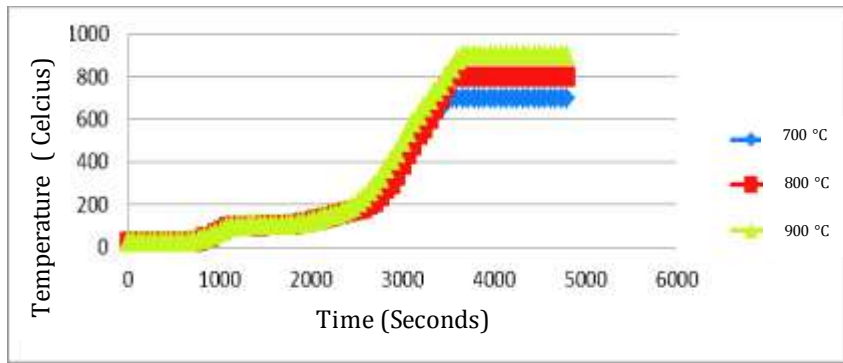
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Fig. 3. Profile of heating rate of gasification process of tobacco leaf waste at various temperature variations with time.

In this evaporation process, the temperature tends to be constant until the end of the gasification process there is no significant increase. Furthermore, at 2700-3100 seconds indicates a temperature range of 250-529 °C, indicating that this condition has entered the pyrolysis zone. Pyrolysis or devolatilization is also referred to as partial gasification, wherein the thermally unstable component referred to in this case is that the lignin present in the tobacco leaf waste will break and evaporate with the other components. Pyrolysis products generally consist of three types, namely light gas (H_2, CO, CO_2, H_2O, and CH_4), tar and char [Basu, 2010].

Time 3200-5000 seconds shows temperature above 550 °C. In this zone indicates the zone of oxidation and reduction, which in this zone occurs exothermically.

reaction. The oxidation process is the most important process because this process generates the heat required by the entire reaction that occurs in the gasification reactor, then the gas produced in the oxidation zone will react in the reduction zone to produce a combustible gas

### 4.2 The Effect of Process Time on Gas Result

In the process time gasification process has an influence in generating product gas. The process time taken in this study is 20 minutes with sampling every 5 minutes to observe the result of the gas composition is able to burn throughout the gasification process. The results obtained from the gasification process (H_2, CO, and CH_4) at temperatures of 700, 800, and 900 °C can be seen in figure 4 below.
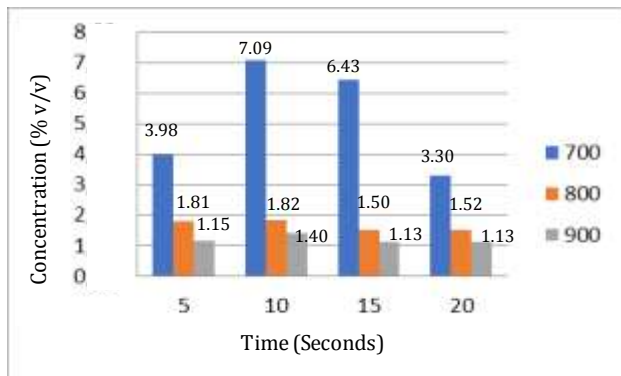
(a)
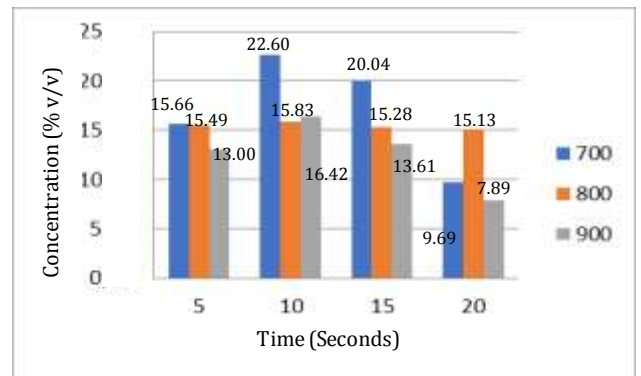
(b)
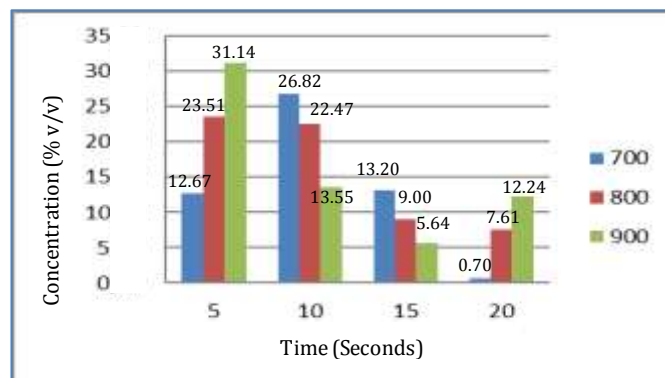
(c)

Fig. 4. The effect of time on the concentration of combustible gas (a) CH_4 (b) CO (c) H_2 at various temperatures.

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In the comparison of the three images above the best results can be seen at 10 minutes at a temperature of 700 °C, almost all components decreased gas composition until 20 minutes. The use of steam can increase the value of carbon conversion to a combustion gas and also as a gasification agent to trigger the whole process through the endothermic reaction of carbon vapor to form hydrogen and carbon monoxide during the course of gasification. The 10 minutes time is the peak of carbon conversion, indicating that the addition of steam can increase carbon conversion in the gasification process. At Fig. 4. Shows as the carbon conversion after the 10 minute shows a decrease. The longer the reaction time of the product gas will increase with time. carbon conversion is affected by the amount of fuel (feed) that is still available. The decrease in gas production (CO, H_2, and CH_4) is due to the fuel used decreases during the gasification process takes place.
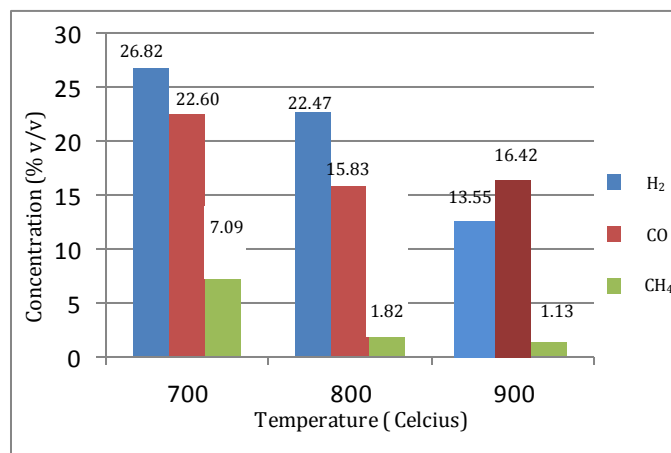
Reduced fuel (feed) leads the less the feed converted to the gas product, so the concentration of gas (CO, H_2 and CH_4) produced will decrease until 20 minutes. The decrease of syngas concentration (CO, H_2 and CH_4) is because the fuel (feed) used decreases during the gasification process. Reduced fuel (feed) results in fewer feeds being converted into syngas (CO, H_2 and CH_4) resulting in lower concentration of syngas (CO, H_2 and CH_4) [Suhendi, *et al*, 2016]

4.3 The Effect of Temperature on Gas Result

Temperature is an important parameter in the gasification process because it affects the chemical reactions that occur. For this purpose, a test of temperature variation of 700 °C, 800 °C, 900 °C with the results obtained from the gasification process (H_2, CO, and CH_4) is presented in Fig.5.

Fig. 5. The effect of gasification temperature on H_2, CO and CH_4 gas component concentration at 10 minutes gasification process with the variation of achieved temperature is made constant

At a temperature of 700 °C can be seen the trend of CO production more than the temperature of 800 oC and 900 °C. At temperature 700 °C obtained CO 22.60% vol, then decreased to 15.83% at a temperature of 800 °C and then increased to 16.42% at 900 °C. This occurs because at lower temperatures the more dominant reaction is the boudouard reaction, resulting in higher CO production compared to high temperatures [Almeida, 2017], whereas for the H2 component there is a decrease of 26.82 vol% at a temperature of 700 °C to 22.47% vol and 13.55 vol% for temperatures of 800 oC and 900 °C, higher temperatures contribute to a higher hydrogen content in gas yields, increasing temperatures provide energy for hydrogen production endothermic reactions, thereby increasing the hydrogen content of the gas yield. At high temperatures, the water-gas reaction has a greater contribution than the Boudouard reaction. In addition to high temperatures water-gas shift reaction (CO + H_2O → CO_2 and H_2) causes an increase in H2 concentration in the producer gas [Mahishi, 2007] and higher temperatures tend to favor steam reform of methane to CO and H_2 [Kumar, 2009].
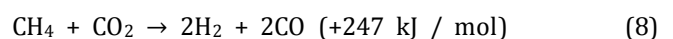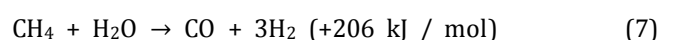
But the results of this study do not match the theory, because the temperature is too high does not match the fuel mass so that the fuel runs out in line with the increase in temperature gasifier and duration

Gasification reaction time, therefore the gas composition obtained is decreasing. The CH_4 concentration obtained at temperatures of 800 °C and 900 °C was 1.82 % vol and 1.40 % vol lower than the methane composition at 700 °C at 7.09 vol%, CH_4 concentration decreased with increasing temperature, occurs because higher temperatures tend to favor methane steam reform that decreases methane concentrations and increases the concentration of hydrogen and carbon monoxide [Wu, *et al*, 2006]. Here's the reaction:

$$CH_4 + H_2O \rightarrow CO + 3H_2 \ (+206 \ kJ \ / \ mol) \tag{7}$$

$$CH_4 + CO_2 \rightarrow 2H_2 + 2CO \ (+247 \ kJ \ / \ mol) \tag{8}$$

CH_4 results in this study were lower than experiments performed by Kumar who got a CH_4 result of 23%

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because the temperature of the study was higher than the temperature performed by the kumar.

4.4 The Effect of K₂CO₃ Catalyst on Gas Result

The development of catalytic gasification is driven by the need for reforming tars, when the product gas passes through the catalyst particles, tar or hydrocarbons can be condensed and reformed on the surface of the catalyst with vapor or carbon dioxide, resulting in additional hydrogen and carbon monoxide.
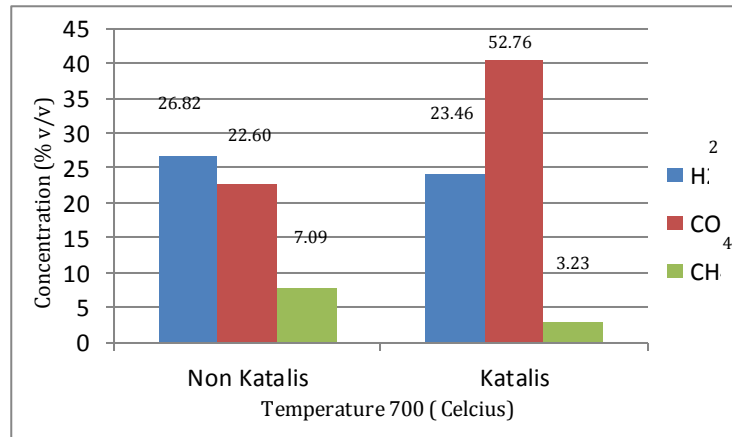

Fig. 6. Effect of adding K2CO3 catalyst to the concentration of H_2, CO and CH_4 gas components at 10 mins gasification process with constant temperature of 700°C.

In this research, the addition of catalyst at temperature of 700 °C to find out the product of gas composition produced, with the addition of K_2CO_3 catalyst obtained H_2 content of 23.46% vol and CO of 52.76% vol at 10 minutes. This gas content increases if compared with the gas yield at the temperature of 700 °C.

without using a catalyst of only 22.82 vol% for CO and 26.82 vol for H_2. this occurred because the addition of the catalyst was able to accelerate the reaction so that the carbon conversion was achieved maximally, Yang Zhang conducted a study of the addition of K_2CO_3

catalyst to the gasification process and gained carbon conversion from 60% to 80% at 600 °C [Mahishi, 2007]. the catalytic mechanism of K_2CO_3 occurring is explained by the reaction as follows:

$$K_2CO_3 + 2C \rightarrow 2K + 3CO \qquad (9)$$

$$2K + 2H_2O \rightarrow 2KOH + 2H_2 \qquad (10)$$
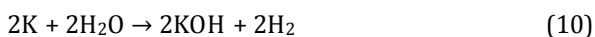
The decomposed catalyst will react with the CO_2 that has formed in the gasification process and return to K_2CO_3

$$2KOH + CO_2 \rightarrow K_2CO_3 + H_2O \qquad (11)$$
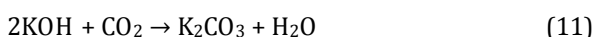
From the explanation of the reaction mechanism can be known formation of 3 moles of CO followed by the formation of 2 moles of H_2. Therefore, with the addition of this K_2CO_3 catalyst can increase the content of CO and H_2 components. However, it can be seen in Fig. 6. that with the addition of catalyst can reduce the content of H_2, this is probably because H_2O does not react with

catalyst so that H_2 gas concentration is not formed perfectly. In addition K_2CO_3 catalysts are included into alkali metal types, this type of catalyst can support the reform of methane into hydrocarbons.

$$CH_4 + H_2O \rightarrow CO + 3H_2 \ (+206 \ kJ \ / \ mol) \qquad (12)$$
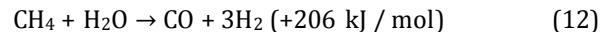
Therefore, the resulting CH_4 decreased compared to a temperature of 700 °C without a catalyst of 3.07 vol%.

5. CONCLUSION

Based on the data and analysis of updraft gasification result with temperature variable and waste catalyst used by batch method to achieve optimum condition of gas process to gas produced, it is concluded that Temperature is very sensitive production of gas product, temperature 700 °C produce gas composition used larger, The best time in the 10th minute, this is evidenced in the graph of the variables decreasing after the 10 minutes, the effect of the K_2CO_3 catalyst produces more gas composition, especially for the increase of CO concentration gas with no catalyst.

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