

The Utilization of Fixed Bed Coal Gasification By-Products to Produce Combustible Gas by Auto-thermal Process

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Abstract— Fixed bed gasification of coal generate some by-products, such as tar and coal powder. Coal powder obtained because the coal feed for fixed bed gasification reactor must have a size above 20 mm. Meanwhile, tar is a high molecular weight hydrocarbon compounds that will condense at low temperatures, which cause clogging and blocking of pipes. In this study, experiment on combustible gas generation from co-gasification of tar and coal powder was conducted in an auto-thermal reactor to determine the temperature of the process, combustible gas composition, and efficiency of the process. From calculation and experiments about oxidizing reactor operation, 20 kg/hr of tar was more promising to operate and can reach the optimal temperature which was 1900°C. The energy from oxidizing reactor used for the reduction reaction of tar and pyrolysis of coal powder and produce combustible yield gas. The coal powder that can be conversion was about 14.4 kg/hr and produce approximately 84.52 kg/hr combustible gas. The calorific value of combustible yield gas amounted to 783.62 Cal/g. Combustible yield gas has advantages levels of hydrogen gas (H₂) as high as 19.2%, which is already exceeding the levels of hydrogen gas from coal gasification is only <10%. Cold gas efficiency (CGE) has a value which was still low at 26.31%. It is caused by two factors, namely the calorific value fuel gas produced was still low and the remainder of the conversion of coal powder were still mostly in the form of charcoal.

Keywords— tar reforming; combustible gas; carbon conversion; cold gas efficiency.

I. INTRODUCTION

Coal is an alternative energy source than petroleum, coal consumption in the country is the biggest to the needs of power plants, while for other industries such as cement, steel, and small industry is still relatively small. Indonesian coal production and consumption will continue to increase in line with economic growth and energy needs [1]. Coal can be converted into combustible gas through gasification process. Besides generate combustible gas, fixed bed coal gasification can also produce some by-products, such as ash, tar, and coal powder. Coal powder obtained because the coal feed for fixed bed gasification reactor must have a size above 20 mm. Meanwhile, tar is a high molecular weight hydrocarbon compounds [2],[3],[4]. Tar will condense at low temperatures, which cause clogging and blocking of pipes, valves, filters, engine and fuel cell stack. Tar can be removed by physical processes that use scrubbers, carbon adsorption and electrostatic precipitator [5]. Increased temperature gasification is a promising approach to reduce the formation of tar [2]. Houben *et al.* [3] carried out tar reduction through partial combustion burner of a

combustible gas experiment. In his study, naphthalene is added as a model tar component. For higher air/fuel ratio values, the total tar content slightly decreases. At lower air fuel ratio and higher hydrogen concentrations, the tar content strongly decreases. It is found that the partial combustion burner reduces the tar content of the gas with over 90% by cracking with an air/fuel ratio of 0.2.

Many studies show that partial oxidation temperature of tar range from 600 to 1300°C. Stravinkas *et al.* [6] reported that tar destruction was performed in a fixed bed reactor at temperatures from 700 °C to 900 °C, with the steam-to-carbon ratio (H₂O/C) from 0 to 1, air equivalence ratio (ER) from 0 to 0.5 at a constant space velocity and the initial concentration of tar 21.1 ± 1.89 g/m³. The thermal destruction of tar was found to be most intensive in terms of both tar destruction and hydrogen gas production when using steam reforming at 900°C and H₂O/C = 1. Under these conditions, an almost complete conversion of tar was achieved, with benzene as the only remainder at a concentration of 0.019 g/m³. In the experiment of Ma *et al.* [7], a combined system of an oxygen carrier material (OCM) and a catalyst for the partial oxidation of naphthalene (as a model component for tar) will be introduced. OCMs are

able to incorporate oxygen ions into the crystal lattice. The oxygen stoichiometry of the ceramic material depends significantly on oxygen partial pressure and temperature. The properties of the OCMs were examined in a long-term four-cycle experiment. In the first cycle, the pre-oxidation phase occurs. In this phase, the OCM was fully oxidized during heating up to 900°C with 25 cm³min⁻¹ air flow. The OCM was then heated up to 800°C in this reducing atmosphere. Because of the low O₂ partial pressure, the oxygen stored in the OCM would then be released in this reduction phase. In combination with the combined catalyst and oxygen carrier system, the use of H₂S sorbent further enhanced the performance of the catalyst. In conclusion, the suitable oxygen carrier materials, catalyst and sour gas sorbent could be identified, which in combination enable the proposed tar removal process.

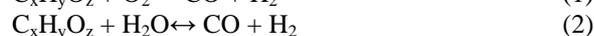
Ahrenfeldt *et al.* [8] found that practically all phenol is converted at temperatures above 950°C. The resulting polycyclic aromatic hydrocarbons (PAH) tar compounds are readily converted in the subsequent char-bed of the Two Stage gasification process and the partial oxidation process thus contributes directly as well as indirectly to the overall tar destruction. A high temperature and excess air ratios contribute positively to the direct tar destruction and a high moisture content of the biomass enhances the decomposition of phenol and inhibits the formation of naphthalene. Ma and Muller [9] reported that four perovskite-type oxide materials and NiO were tested regarding their catalytic activity for partial oxidation of naphthalene as model tar from biomass gasification. Using these catalysts, at least 60% of naphthalene was converted above 600°C. NiO, which is reduced to Ni under gasification conditions, showed the highest catalytic activity of all tested materials with about 95% conversion at 600°C.

Partial oxidation over a wide temperature range from 1100 to 1400°C was studied by Tsuboi *et al.* [10]. They were investigated regenerative reforming of light and heavy tars contained in syngas by steam gasification. In a first step, basic design parameters of the reformer such as an appropriate size of the reactor and amount of oxidant were determined by using numerical predictions. The results showed that the appropriate average temperature in the reactor, necessary minimal residence time and oxygen flow rate are 1300°C, 4 s and 12% of the syngas volume, respectively. Experiments with the new regenerative tar reformer proved stable operation and reforming efficiency exceeding 99% at an oxygen flow-rate of 11.3%. This result also proved that the concept of regenerative reformer yields higher system efficiency because the same high reforming efficiency as obtained with a conventional reformer could be achieved with 30% less oxygen consumption. In the comparison of two oxygen nozzle designs, four holes yield higher efficiencies (η_{tar}) than a one-hole nozzle due to better mixing of syngas and oxygen. It was found that the formation of a high-temperature zone has a strong effect on high reforming efficiency. Zhao *et al.* [11] also investigated the tar components, tar conversion rates, a physical and chemical structure of biochar after reaction at the second stage were sampled and analyzed. Results showed that at 700°C, the coupling of char and oxygen could result in the significant improvement of tar conversion rate (89.32%)

than both two separated method (85.1% and 86.14%). At 900°C, the synergy effect could reach the highest conversion rate of 95.84%.

Van der Hoeven *et al.* [12] also investigated partial producer gas combustion for tar reduction. The study showed that the influence of ambient gas such as hydrogen on thermal tar conversion can probably be derived from the chemical balance of the reactions taking place. The addition of a certain ambient gas can generate driving forces which can direct or redirect species balance of reactions. The results showed that a rising hydrogen content is always beneficial for increased tar cracking, by increased reaction rates, increased free radical production, and increased radical residence times. Therefore, to obtain best tar cracking by partial product gas combustion, the product gas should have a high hydrogen content.

Based on the few studies that have been conducted, tar was converted using a reagent mixture of oxygen-steam or air-steam causing partial combustion of the chemical reaction as follows:



The reaction is exothermic and endothermic, so overall not require additional heat from outside the system. Autothermal conversion technology is known as partial oxidation (partial oxidation, POX). Comparison of different pitch conversion technology can be seen in Fig.1.

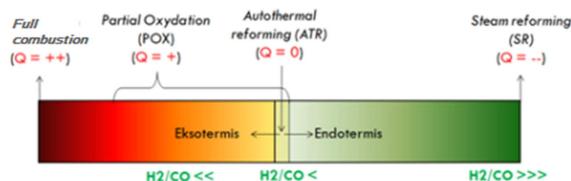


Fig 1. Conversion technology into a combustible gas [13]

However, the existing processes have not resolved the problem of another by-product which is coal powder. To overcome these problems, an innovative gasification process that considers the tar oxidation and coal powder gasification stages in the reactor is proposed. The tar oxidation stage is designed for tar combustion fed with air. As the temperature rises to the desired temperature, tar added to the partial oxidation reactor and the coal powder was added to the reforming reactor to serve as gasification raw materials during the process. The gasification-required heat was provided by the oxidation/combustion reactor. This gasification technology operates in the different reactor; thus, all the heat of coal combustion can supply the reforming/gasification stage, which is better for tar cracking and can effectively prevent the sintering of raw materials in the gasification process. Based on this technology, on combustible gas generation from co-gasification of tar and coal powder was conducted in an auto-thermal reactor to determine the temperature of the process, combustible gas composition, and efficiency of the process.

II. MATERIAL AND METHOD

Experiments were performed at atmospheric pressure, auto-thermal, and gasification system. Fig. 1 presents the schematic of the gasification system, which mainly includes

an auto-thermal gasifier, two separated feeders (tar and coal), a high-temperature cyclone separator, a temperature controller, a tube type heat exchanger, and a chimney. Process flow diagram of research equipment shown in Fig.2.

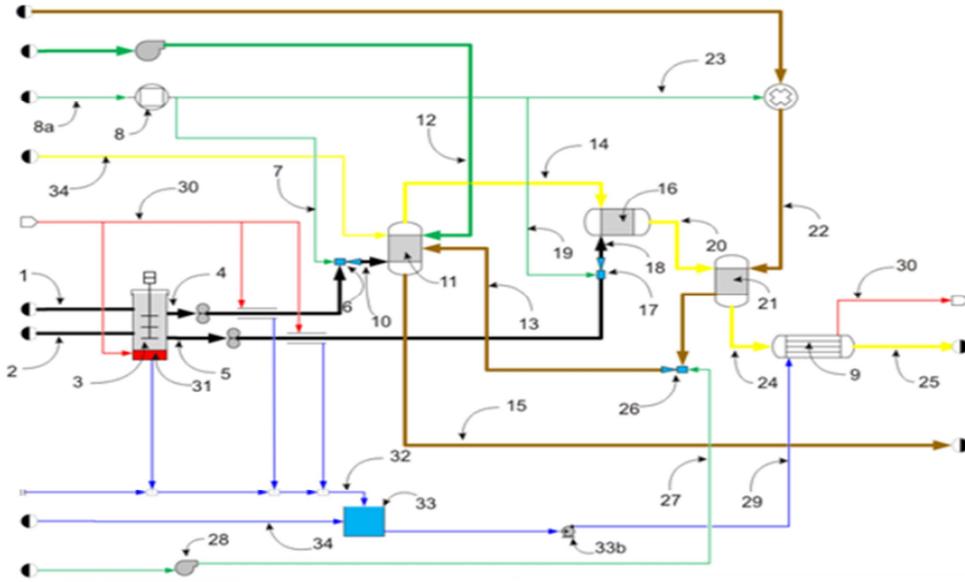


Fig 2. Process flow diagram of research equipment.

The flow rate of tar to the oxidation chamber set at 15 L / hr. The furnace process consists of seven stages: the heating tar as fuel combustion (1) and (2) in the tank (3) to decrease the viscosity of tar so it is easy to flow into research equipment; tar combustion reaction (4) and coal powder charcoal recycle (13) with air in an oxidizing reactor (11) generates an oxidizing gas (14) and liquid ash or slag (15); reduction reaction between the oxidizing gas (14) with tar feed (5) in the reductant reactor (16) gas into a reductant gas (20); catalytic reactions reforms between reductant gas (20) with the coal powder (22), which consists of the reaction of pyrolysis of coal powder, catalytic reactions charcoal for cracking long chain hydrocarbon and the catalytic reaction of alkalis for the reduction reaction of coal powder into producer gas (24) and coal powder (13); coal powder charcoal-recycle to the oxidant (11) and as fuel for the combustion reaction; gas producer (24) is cooled by water cooling medium (29) in the waste heat boiler (9), in order to obtain steam (30); steam (30) used for the heating medium. In the gasification stage, the sample of the product gas is collected from the bell-type gas holder with a gas bag and analyzed for major components (H_2 , CO , CO_2 , and CH_4).

Data processing experiments were conducted to determine the performance of the tar conversion process into combustible yield gas. Processing data includes into combustible gas composition analysis, calculation of mass balance, energy balance, carbon conversion and cold gas efficiency (CGE). The composition of fuel gas conducted to determine the composition of impervious gas fuels (combustible gasses) is H_2 , CO , CH_4 and C_{2+} and unreceptive fuel gas composition (noncombustible gasses), such as CO_2 and N_2 . Analysis of fuel gas using gas

chromatography. Mass balance calculation was conducted by calculating the flow rate of feed (tar, air and coal powder) and the calculation of the flow rate of the product (fuel gas and charcoal). Energy balance calculation was done to determine the chemical energy content in the fuel gas product. Carbon conversion was used to determine the amount of carbon from the feed (tar and coal) which converted into a fuel gas. While the cold gas efficiency (CGE) was used to determine the chemical energy in the fuel gas produced in the process of the chemical energy contained in the feed (tar and coal).

III. RESULTS AND DISCUSSION

A. Tar Characteristics

In this research, tar samples used were obtained from a ceramic factory PT. SANGO in Semarang, Indonesia. Laboratory analysis of the tar can be seen in Table I. The results of the laboratory analysis showed that the water content of tar was very small, which was 2%, eliminating the need for separated water content.

Although tar has a low pour point, but still contains a high carbon residue, which was 10% weight, and high levels of sedimentation which was 0.36%. This causes tar susceptible for coagulation when it flowed in the pipeline. One way to overcome it was by warming the tar up to 60-65°C, then filtered using a filter with a size of 100 mesh.

After tar fed into the tank, then tar was heated using steam heated by setting the temperature 88 - 90 ° C. This setup is done automatically using the electronic control valve regulated by a micro-controller on the control panel. Along the heating process in the tank, conducted warm up pipe flow towards the nozzle. Steam was used from the

start-up boiler which uses LPG fuel. Start up the boiler will be shut down operations if the steam generated by Gas Cooler has been fulfilled. Gas Cooler products utilizing sensible heat gas to generate steam.

TABLE I
PROXIMATE AND ULTIMATE ANALYSIS OF TAR

Parameter	Unit	Value	Method
Calorific value	Cal/g	8,650.72	ASTM D.4809
Kinetic viscosity 100°C	cSt	12.25	ASTM D.445
Ash content	% w (weight)	0.069	ASTM D. 482
Pour point	°C	30	ASTM D.97
Flash point	°C	136.5	ASTM D.93
C content	%w	79.34	ASTM D.5291
H content	%w	7.87	ASTM D.5291
N content	%w	1.47	ASTM D.5291
O content	%w	10.36	ASTM D.5291
S content	%w	0.34	ASTM D.4294
Water content	%w	2.0	ASTM D.95
Carbon residue	%w	10.44	ASTM D.4530

B. Coal Powder Characteristics

Coal powder that used in this research was from finely unused coal in a fixed bed gasifier in Palimanan, Indonesia. Characteristics of coal can be seen in Table II. The coal powder was categorized in sub-bituminous coal. In the fixed bed gasifier, this coal powder was a byproduct because of the coal used was nut size which is over 2 cm.

TABLE II
PROXIMATE AND ULTIMATE ANALYSIS OF COAL POWDER

Parameter	Unit	Value	Method
Proximate			
Moisture	%, adb	14.81	ASTM D. 3173
Ash	%, adb	2.32	ASTM D. 3174
Volatile matter	%, adb	41.99	ASTM D. 3175
Fixed Carbon	%, adb	40.88	ASTM D.3172
Gross calorific value	Cal/g, adb	5,293	ASTM D.5865
Total sulfur	%, adb	0.25	ASTM D.4239
Ultimate			
Carbon	%, adb	57.1	ASTM D.5373
Hydrogen	%, adb	5.51	ASTM D.5373
Nitrogen	%, adb	0.91	ASTM D.5373
Oxygen	%, adb	3.31	ASTM D.3176
Hardgrove Grindability Index		33	ASTM D.409
Ash fusion temperature (reducing atmosphere)			
Deformation temperature	°C	1,212	ASTM D.1857
Spherical temperature	°C	1,221	ASTM D.1857
Hemisphere temperature	°C	1,224	ASTM D.1857
Flow temperature	°C	1,230	ASTM D.1857
Ash fusion temperature (oxidizing atmosphere)			
Deformation temperature	°C	1,223	ASTM D.1857
Spherical temperature	°C	1,235	ASTM D.1857
Hemisphere temperature	°C	1,239	ASTM D.1857
Flow temperature	°C	1,283	ASTM D.1857

C. Oxidizing Reactor Operation

Oxidation of tar in the oxidizing reactor was conditioned to obtain optimal conditions of heat that can be supplied for tar conversion into fuel gas in reductant reactor. Fig 3.

shows the correlations between the flow rate of air used to burn the tar of temperatures in the oxidizing agents with different tar feed, which were 5, 10, 15, and 20 kg/hr. Air flow rate varied according to the needs of stoichiometric combustion of tar (70-150% stoichiometric). As reference was the stoichiometric air requirement burning 100% of each feed tar, which was 47 m³/hr of air for 5 kg/hr of tar, 94 m³/hr of air for 10 kg/hr of tar, 141 m³/hr of air for 15 kg/hr of tar, and 188 m³/hr of air for 20 kg/hr of tar.

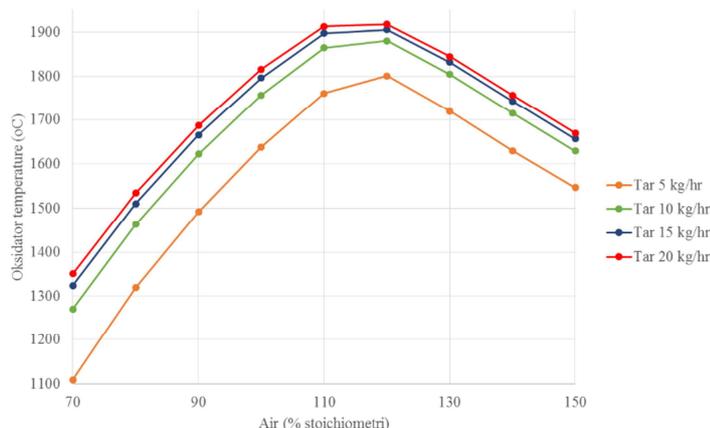


Fig 3. Correlations of the flow rate of air and temperature in the oxidizing reactor.

In Fig 3., it can be seen that the combustion in the oxidizing reactor reached 1,900°C. This condition was obtained when the tar feed at 20 kg/hr with 110-120% stoichiometric air. But after passing 120% stoichiometric air, the heat was generated slowly began to decline. At the time of the burning, tar was able to be burned until 120% stoichiometric air. When air was supplied above 120% stoichiometry, tar that be burned is not available anymore and the heat generated is absorbed by the excess air. The air is represented by the amount of unburned oxygen in the flue gas in which can be seen in Fig 4.

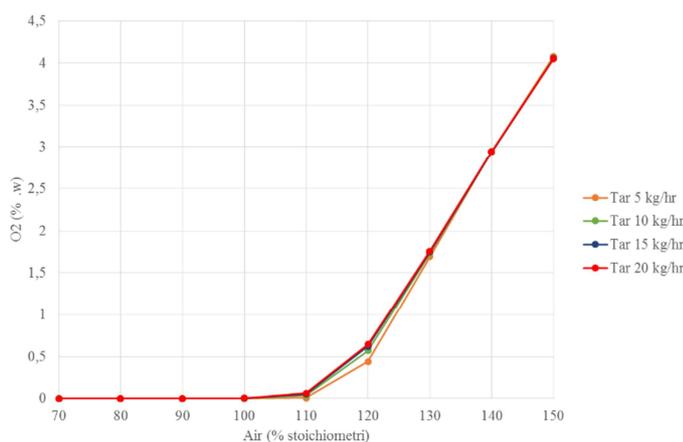


Fig 4. Correlations of the air flow rate and the oxygen content in the flue gas.

The amount of oxygen began to rise drastically after the stoichiometric air passing through 120%. Furthermore, the indicator of the temperature drop in oxidizing reactor was the presence of carbon dioxide (CO₂) as shown in Fig 5.

Complete combustion reaction produces CO_2 and H_2O [14]. The amount of CO_2 reached record levels at 120% stoichiometric air. After passing these conditions, the amount of CO_2 begins to decline.

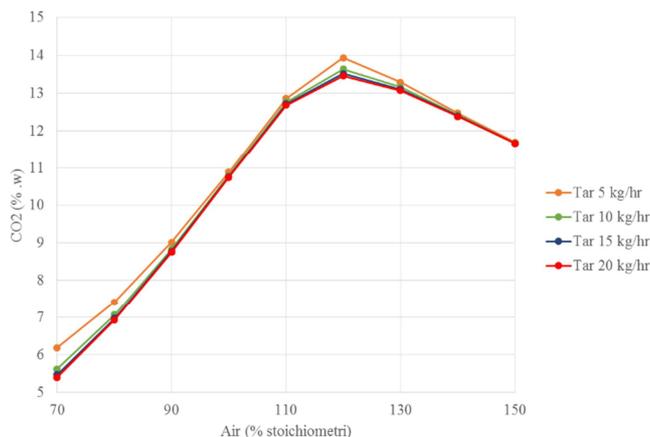


Fig 5. Correlations of the air flow rate and the carbon dioxide content in the flue gas.

The amount of nitrogen dioxide (NO_2), which is the emission on combustion, is also influenced by the presence of O_2 [15]. NO_2 has increased more than 120% when the air stoichiometry, as shown in Fig 6.

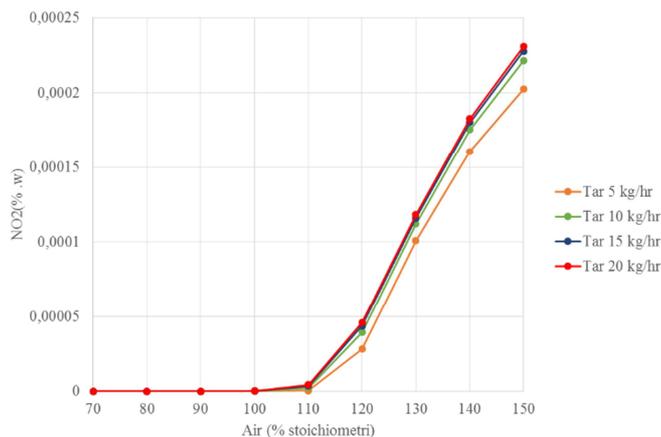


Fig 6. Correlations of the air flow rate and the nitrogen dioxide content in the flue gas.

Nitrogen contained in the fuel can be bound with oxygen to form oxides with varying degrees of oxidation [16]. Inversely with NO_2 , sulfur dioxide (SO_2) in the oxidizing reactor has decreased, it is caused by a drop in temperature in the reactor. When the temperature increases, the formation reaction of SO_2 will be more slowly that decrease the amount of SO_2 , as shown in Fig 7.

On the number of different tar feed, oxidizing reactor temperature and gas content will be almost equal. However, the air flow rate varied according to stoichiometry, so that in the larger tar feed, the reactor conditions become more stable. In Fig 3. and Fig 5., for 5 kg/hr of tar, the process looks more volatile than 20 kg/hr of tar. Therefore the feed of 20 kg/hr of tar was easier to operate and more promising than with smaller amount, in addition to consideration of the optimal temperature that can be reached was 1,900°C.

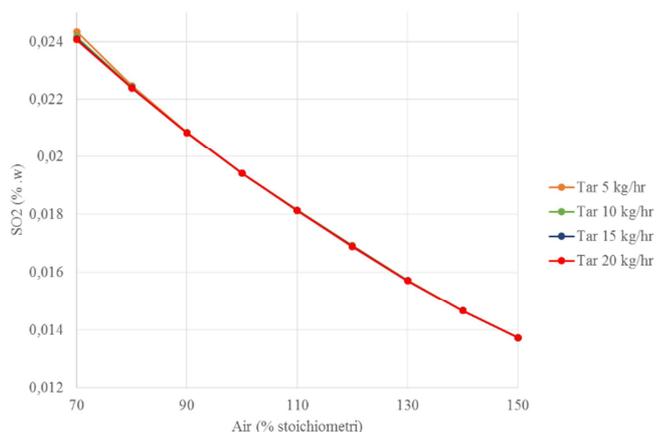


Fig 7. Correlations of the air flow rate and the sulfur dioxide content in the flue gas.

D. Combustible Yield Gas

The calorific value of combustible yield gas amounted to 783.62 Cal/g as shown in Table III. It still can be improved by increasing the gas content of carbon monoxide (CO) and reduce carbon dioxide (CO_2) through the reduction reaction of CO_2 to CO in the reactor reductants. Combustible yield gas has advantages levels of hydrogen gas (H_2) as high as 19.2%, which is already exceeding the levels of hydrogen gas from coal gasification is only <10%. Hydrogen gas is the fuel that is environmentally friendly because it does not produce the greenhouse effect [17]. This is due to feed tar has a chemical composition ratio H/C which is higher than coal.

TABLE III
RESULTS ANALYSIS OF COMBUSTIBLE YIELD GAS

Parameter	Unit	Value
H_2	% vol.	19.2
O_2	% vol.	0
N_2	% vol.	50.80
CH_4	% vol.	0.5
CO	% vol.	8
CO_2	% vol.	19.3
C_2+	% vol.	2.2
Total	% vol.	100
Calorific value	Cal/g	783.62

E. Efficiency

Calculation of efficiency conversion process tar and coal powder into gas fuel can be seen in Table IV. Coal powder was fed into the reformer reactor and will be pyrolyzed into fuel gas and charcoal. The gas flow rate in the reformer reactor occur more quickly, so coal powder will be carried in the gas stream and leave the reformer reactor in a shorter time.

Cold gas efficiency (CGE) has a value which was still low at 26.31% compared to gasification which have efficiency 50-70% [18]. It is caused by two factors, namely the calorific value fuel gas produced was still low and the remainder of the conversion of coal powder were still mostly in the form of charcoal. Table IV showed the coal

powder that was converted into gas, which amounted 14.4 kg/hr.

TABLE IV
COLD GAS EFFICIENCY ANALYSIS OF AUTO-THERMAL CONVERSION PROCESS

Parameter	Inlet		Outlet
	Tar	Coal powder	Combustible gas
Flow (kg/hr)	20	14.4	84.52
Calorific Value (Cal/g)	8,560.72	5,293	783.62
Energy (kCal/hr)	175,545.45	76,219.25	66,228.21
	251,764.75		
Cold Gas Efficiency (%)	26.31		

IV. CONCLUSION

An experiment on combustible gas generation from co-gasification of tar and coal powder was conducted in an auto-thermal reactor to determine the temperature of the process, combustible gas composition, and efficiency of the process. From calculation and experiments about oxidizing reactor operation, 20 kg/hr tar was more promising to operate and can reach the optimal temperature which was 1,900°C. The energy from oxidizing reactor used for the reduction reaction of tar and pyrolysis of coal powder and produce combustible yield gas. Coal powder was fed into the reformer reactor and will be pyrolyzed into fuel gas and charcoal. The coal powder that can be conversion was about 14.4 kg/hr and produce approximately 84.52 kg/hr combustible gas. The calorific value of combustible yield gas amounted to 783.62 Cal/g. It still can be improved by increasing the gas content of carbon monoxide (CO) and reduce carbon dioxide (CO₂) through the reduction reaction of CO₂ to CO in the reactor reductants. Combustible yield gas has advantages levels of hydrogen gas (H₂) as high as 19.2%, which is already exceeding the levels of hydrogen gas from coal gasification is only <10%. Hydrogen gas is the fuel that is environmentally friendly because it does not produce the greenhouse effect. This is due to feed tar has a chemical composition ratio H/C which is higher than coal. Cold gas efficiency (CGE) has a value which was still low at 26.31% compared to gasification which have efficiency 50-70. It is caused by two factors, namely the calorific value fuel gas produced was still low and the remainder of the conversion of coal powder were still mostly in the form of charcoal.

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