EFFECT OF LOW RANK COAL TEMPERATURE AND MOISTURE CONTENT ON SLOW PYROLYSIS PROCESS

PENGARUH SUHU DAN KADAR AIR BATUBARA PERINGKAT RENDAH TERHADAP PROSES PIROLISIS LAMBAT

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ABSTRACT

The government of Indonesia has made policies to increase the added value of coal. It can be completed using technology of slow pyrolysis. This study aimed to increase the added value of coal by analyzing the effect of temperature and water content of the feed on slow pyrolysis products. Water content variation in the feed form of coal was 10.57 wt% and 16.86 wt%. Temperature variations in the slow pyrolysis process were 500, 600, 700, and 800 ºC. Result of this research showed that the low moisture content of coal produced more char in the slow pyrolysis process. Based on the variations of the used temperature, a higher temperature of pyrolysis produced fewer char but its calorific value of coal product was also higher. The optimum operating condition was achieved at 500-600 ºC.

Keywords: slow pyrolysis, water content, temperature, char quality, char energy content.

ABSTRAK

Pemerintah Indonesia telah membuat kebijakan untuk meningkatkan nilai tambah batubara. Hal ini dapat dilakukan dengan menggunakan teknologi pirolisis lambat. Oleh karena itu, tujuan penelitian ini adalah untuk meningkatkan nilai tambah batubara. Penelitian ini dilakukan dengan menganalisis pengaruh suhu dan kadar air umpan terhadap produk pirolisis lambat. Variasi kadar air dalam umpan batubara adalah 10,57 wt% dan 16,86 wt%. Variasi suhu pada proses pirolisis lambat adalah 500, 600, 700, dan 800 °C. Hasil penelitian menunjukkan bahwa batubara dengan kadar air rendah menghasilkan lebih banyak arang pada proses pirolisis lambat. Berdasarkan variasi suhu yang digunakan, semakin tinggi suhu pirolisis semakin sedikit arang yang dihasilkan, tetapi nilai kalor produk batubara lebih tinggi. Kondisi operasi optimum diperoleh pada suhu 500-600 °C.

Kata kunci: pirolisis lemah, kandungan air, suhu, qualitas arang, kandungan energi arang.

INTRODUCTION

Based on the data from the Geological Agency of the Ministry of Energy and Mineral Resources of Indonesia, the total of Indonesia's coal reserves was 26.2 billion tons in 2019. With 461 million tons of coal production in 2018, coal resources amounted to 124.6 billion tons. About 60% of the coal reserves consist of low-rank coal with a caloric content of less than 5,100 kcal/kg. The government of Indonesia has made regulation number 25 in 2018 on the Implementation of Mineral and Coal Mining Activities to increase the added value for mineral and coal commodities.

Increasing the quality of coal can be accomplished through the pyrolysis process. Coal pyrolysis is the first stage in all thermal processes of coal utilization such as combustion, carbonization, gasification and thermal enhancement in limited air conditions (Gavalas, 1982). The coal pyrolysis conditions

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will influence the char characteristics, and such the characteristics are closely related to the char gasification reactivity (Wang *et al.*, 2016). Pyrolysis is a thermochemical decomposition of biomass into a range of useful products, either in the total absence of oxidizing agents or with a limited supply that does not permit gasification to an appreciable extent (Basu, 2013). Two processes occur competitively when coal is heated. One is the depolymerization process at which gas, water vapor, and tar are formed. The other is the condensation or depolymerization process, which leads to char formation. In these competitive processes, a great number of pyrolytic reactions occur (Seo *et al.*, 2011).

The suitable type of pyrolysis to be developed is slow pyrolysis because it can improve the quality of coal and also produce coal tar as the by-product that can be processed further into synthetic crude and coke oven gas (COG). Slow pyrolysis is pyrolysis that takes place rapidly at temperatures in the range 500 - 950 °C with a heating rate of around 10 until 200 °C/s and residence time in the range of 0.5 – 10 s (Babu, 2008). Slow pyrolysis is able to improve coal quality by improving its proximate, ultimate, and heating value characteristics. The characteristic calorific value increases because the O-C ratio and the H-C ratio decrease during the slow pyrolysis process. When the ratio of hydrogen to carbon (H/C) decreases, the heating value of coal increases (Basu, 2013). Increases in the quality of coal will increase the price of coal.

The characteristics of the solid product from the slow pyrolysis process (high-quality coal) were affected by the parameters of the slow pyrolysis process. Some of the parameters that affect the slow pyrolysis process are the temperature, pressure, duration, heating rate of the slow pyrolysis process, the moisture content of the coal, the size of the coal particles, the height of the reactor bed and the nitrogen gas flow rate. Several researchers experienced the pyrolysis process by producing a low pyrolysis process temperature range between 200 - 300 °C (Tumuluru *et al.*, 2011; Rousset *et al.*, 2012). The result of their coal pyrolysis research showed that the coal pyrolysis process was able to produce coal that has a higher heating value than the feed. The purpose of this study was to determine the effect of slow pyrolysis temperature and coal moisture content on coal product quality.

METHOD

Material

The equipment used in this study were a coal screw feeder, fixed bed reactor, a heat exchanger (HE)/condenser, and a coal gas analyzer. The fixed bed reactor was made of quartz glass and equipped with electric heaters. The condenser cools and condenses liquid products (water and tar) carried in non-condensable gas (NCG) products. The test equipment configuration can be seen in Figure 1. The materials used in this study consisted of coal, nitrogen gas (N_2), and anisole (methoxybenze).

Method

There were two variables in this study. The first variable was the coal variable condition: 20 gram/hour of coal and -32+42 mesh of coal size. The second variable was changed variable that consisted of reactor temperatures (500, 600, 700, and 800 ºC) and moisture of coal (10.57 wt. % air dried basis (adb) of Coal A and 16.86 wt. % adb of Coal B). Experimental procedures performed in this study were preparation and testing, the yield of pyrolysis products measurement, the quality of char determination, and material balance calculation. In the preparation and testing stage, the pyrolysis process was carried out in a continuous scheme. Twenty grams of +32 mesh coal were put into the reactor for 1 hour and tested according to the reactor temperature variation (500, 600, 700, and 800 ºC).

The experiment was started by streaming a nitrogen gas into the pyrolysis equipment. The nitrogen gas (N_2) was streamed to the reactor and gas cooling equipment to remove the trapped air. The nitrogen gas flow rate was maintained at 24 mL/minute. Gas products flowed through the top of the reactor and then cooled through a condenser. The nitrogen gas was also used as a carrier gas during the experiment. When the experiment occurred, the used nitrogen flow rate was 0.05 mL/min. The gas out of the condenser was analyzed with a gas analyzer.

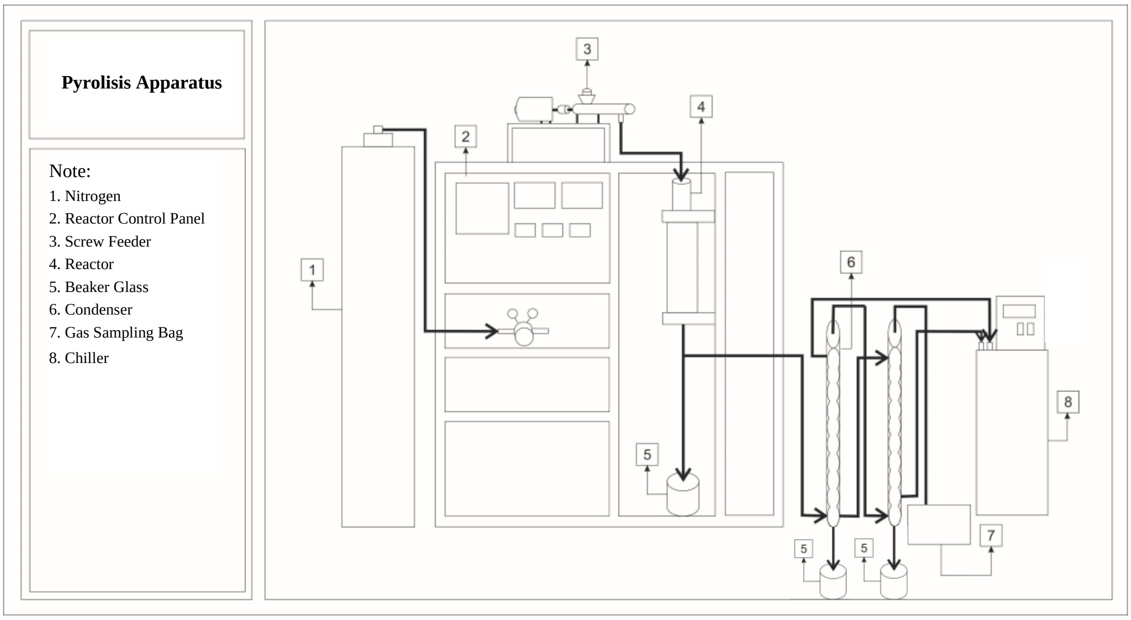

Figure 1. Test equipment configuration of pyrolysis process

The pyrolysis reactor was set at 10 ºC/minute until the temperature was achieved. A 20 gram/hour of coal was put into the reactor. The gas product flowed out from the top of the reactor and then streamed into a condenser equipped with cooling water circulation (5-10 °C). The components of pyrolysis gas were condensed and separated from non-condensable gas (NCG). These liquids were collected in a beaker glass (tar container). NCG was collected in a sampling bag. The NCG in the sampling bag was divided into 4 pieces of impinger. The first 3 units of impinger were containing 60 mL of anisole and the last impinger was containing activated carbon to ensure all tar was removed from NCG. The clean NCG was streamed to a coal gas analyzer and gas composition analysis was done. The composition of the NCG measured was CO, CO_2, H_2, CH_4, O_2, CnHm, and N_2. Methane is the dominant and also the most important aliphatic product of coal pyrolysis. Coal pyrolysis only produces a small number of aromatic products, less than the amounts of H_2O, CO, CO_2, H_2 and CH_4 (Wang *et al.*, 2015).

After the experiment, the yield measurement of pyrolysis products was carried out to obtain the values of char, NCG, and tar. Char and tar were measured by analytical balance. NCG was measured by difference. The proximate, ultimate, and calorific values of char were analyzed with the ASTM method. Heat and material balance was obtained by calculation. A 100 kg of coal was used as the basis for the calculation. The energy content of coal was obtained from the multiplication of 100 kg with the calorific value of coal. The mass of char was obtained from the multiplication of 100 kg with %char. The energy content of char was obtained from the multiplication of mass of char with the calorific value of char. While energy in char was obtained from the division of energy content of char with the energy content of coal.

RESULTS AND DISCUSSION

Effect of Moisture Content and Temperature

The mass balance products of Coal A and Coal B pyrolysis at each temperature were shown in Figure 2. The results showed that the yield of char decreased with increasing pyrolysis temperature, while on the other hand, tar and NCG increased. This result occurs due to the loss of moisture and volatility content in char. Coal can be slowly oxidized even at temperatures lower than 100 °C and the heat is slowly released. The unstable bridge bond on the outer alkyl side chain of coal molecules is broken to form a free radical group, and as the temperature rises, the free radical group starts to be active and the oxidation reaction rate increases. Meanwhile, the evaporation of moisture in coal absorbs heat at this stage. For the heating process of coal in the air atmosphere, both slow oxidation and moisture evaporation happen. For the coal sample with high moisture content, the heat adsorbed is greater than that of the released one (Wang *et al.*, 2020).

Coal A has a lower moisture content than Coal B. Therefore, at the same energy, less char was produced from Coal B than Coal A because the energy was used to evaporate water. This was also indicated by the more liquid product from Coal B than Coal A. The volatile (tar and NCG) yield increases rapidly at low temperatures and then becomes gentler at high temperatures. A peak appears in the tar yield with increasing temperature, resulting from the competition between tar cracking and forming rates (Yi, Feng and Li, 2019).

Char yield tends to flat from 500 to 600 ºC then a significant decrease in char yield occurred from 700 to 800 ºC. This showed that the optimal temperature of coal pyrolysis was 600 ºC for Coal A and Coal B. Increasing the temperature above 600 ºC requires more energy while the resulting char yield was not too different. The yield of char produced at 500 ºC was higher than the yield of char at 600, 700, and 800 ºC. Pyrolysis temperature is the most influential factor among the pyrolysis parameters (Qian *et al.*, 2012).

Char Quality

Pyrolysis is a decomposition process that occurs when volatiles is driven out from hydrocarbon material under heating conditions. The higher the pyrolysis temperature, the produced lower the volatile matter (Table 1 and 2). The ratio of a sharp decrease in volatile matter occurred at temperatures of 500 - 600 ºC for Coal A and Coal B. This will lead to the increased carbon and fixed ash.

The carbon content in char has increased after the pyrolysis process. The carbon content of Coal A before the pyrolysis process was 42.86%, increased to 57.56% at 500 °C, 66.29% at 600 °C, 80.17% at 700 °C, and 79.56% at 800 °C after pyrolysis. The carbon content of Coal B before the pyrolysis process was 38.74%, increased to 65.42% at 500 °C, 74.2% at 600 °C, 79.93% at 700 °C, and 78.23% at 800 °C after pyrolysis.

The carbon content in char increased with increasing pyrolysis temperature, then decreased at 800 ºC because the char which were produced at high pyrolysis temperature contains low volatile matter and more carbonized carbon. Therefore, high carbon products were obtained by increasing the pyrolysis temperature. The results showed that the char content of H, O, N, and S decreased with increasing temperature (Table 1 and 2). The result was caused by pyrolytic processes, which result in the loss of surface functional groups containing O and H (Al-Wabel *et al.*, 2013; Usman *et al.*, 2015). These light molecules were released as volatiles. The major constituents of volatiles were combustible gases like hydrocarbon, hydrogen, carbon monoxide, hydrogen sulfide. When pyrolysis temperature rise, more chemical bonds in coal were broken, more light gases were released as volatiles (Li *et al.*, 2017).

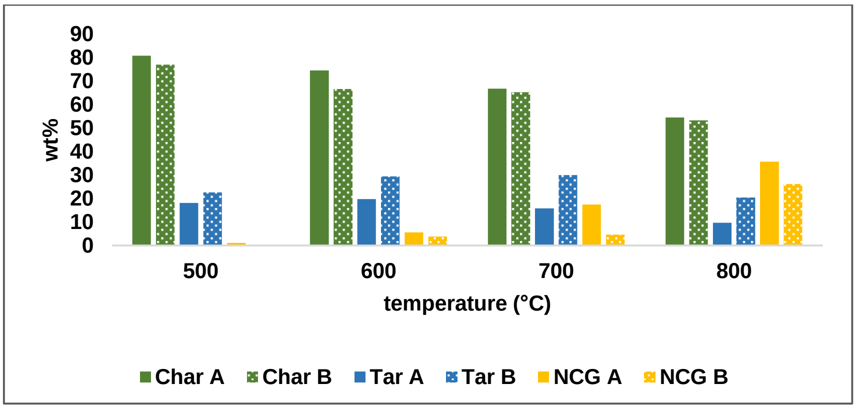

Figure 2. Material balance of slow pyrolysis

An increase in pyrolysis temperature between 500-700 ºC increased the calorific value of char sharply (Table 1 and 2). This was caused by the increase of fixed carbon at 500-700 °C of pyrolysis temperature. Fixed carbon contained a large energy density. The higher the fixed carbon, the higher the calorific value of char (Anupam *et al.*, 2016).

Char Energy Content

The energy content of char to coal resulted from the calculation of mass and energy balance were shown in Tables 3 and 4. Char energy content of in pyrolysis temperatures of 500, 600, 700, and 800 ºC were 95.75, 95.63, 84.11, 65.07% for Coal A and 92.39, 86.12, 84.63, 66.86% for Coal B. From that result, the recommended pyrolysis temperature was 500-600 ºC. The remaining energy was found in other pyrolysis products (tar and NCG). Pyrolysis products that increase with the increase of pyrolysis temperature were tar and NCG as shown in Figure 2. The mixture of tar and NCG was known as a volatile matter (VM). VM has high energy content and will be able to utilize further (Nurhadi *et al.*, 2021). The produced char due to its high carbon content can be used for metallurgical purposes (PCI, pulverized coal injection) and also for mercury removal in flue gases (PAC, powdered activated carbon/coke) (Richter *et al.*, 2017).

Table 1. Properties of coal A

Parameter	Unit	Coal A	A Char			
			500°C	600°C	700°C	800°C
Proximate						
Moisture	%adb	10.57	5.5	3.54	7.14	10
Fixed carbon	%adb	42.86	66.29	77.34	80.17	79.56
Volatile matter	%adb	44.05	24.30	14.48	7.88	5.38
Ash	%adb	2.49	3.91	4.64	4.66	5.06
Ultimate						
Carbon	%adb	60.31	72.34	80.40	80.95	79.43
Hydrogen	%adb	5.24	3.69	2.96	2.58	2.21
Nitrogen	%adb	1.06	1.38	1.47	1.19	0.83
Sulphur	%adb	0.22	0.2	0.26	0.27	0.28
Oxygen	%adb	30.68	18.48	10.27	10.35	12.19
Calorific Value	kcal/kg, %adb	5,688	6,738	7,298	7,166	6,784

Table 2. Properties of coal B

Parameter	Unit	Coal B	B Char			
			500°C	600°C	700°C	800°C
Proximate						
Moisture	%adb	16.86	6.47	6.3	7.59	10.78
Fixed carbon	%adb	38.74	65.42	74.20	79.93	78.23
Volatile matter	%adb	42.66	24.49	14.64	8.03	5.84
Ash	%adb	1.74	3.62	4.86	4.60	5.15
Ultimate						
Carbon	%adb	57.78	71.74	77.96	80.63	78.97
Hydrogen	%adb	6.05	3.91	3.21	2.65	2.28
Nitrogen	%adb	0.90	1.43	1.46	1.14	0.86
Sulphur	%adb	0.18	0.20	0.24	0.26	0.28
Oxygen	%adb	33.35	19.1	12.27	10.72	12.46
Calorific Value	kcal/kg, %adb	5,498	6,591	7,098	7,116	6,738

Table 3. Coal A char analysis

Parameter	Unit	Coal A	A Char			
			500°C	600°C	700°C	800°C
Weight	%	100	80.83	74.54	66.77	54.56
Calorific value	kcal/kg	5,688	6,738	7,298	7,166	6,784
Energy balance	kcal	568,800	544,599	543,956	478,438	370,101
Char energy content to coal	%	100	95.75	95.63	84.11	65.07

Table 4. Coal B char analysis

Parameter	Unit	Coal B	B Char			
			500°C	600°C	700°C	800°C
Weight	%	100	77.07	66.71	65.39	54.56
Calorific value	kcal/kg	5,498	6,591	7,098	7,116	6,738
Energy balance	kcal	549,800	507,968	473,508	465,280	367,592
Char energy content to coal	%	100	92.39	86.12	84.63	66.86

CONCLUSIONS

Low moisture content coal produced more char in the slow pyrolysis process. A high calorific value of char was obtained in the range temperature of 500 and 600 ºC. The utilization of volatile matter from the slow pyrolysis process needs to be investigated further so the pyrolysis technology can be applied in an integrated scheme at the power plant.

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