Effect of temperature and moisture content of low rank coal to slow pyrolysis process

PENGARUH SUHU DAN KELEMBABAN BATUBARA PERINGKAT RENDAH TERHADAP PROSES PIROLISIS LAMBAT

Slamet Handoko\*, Sapta Rianda, and N Nurhadi

R&D Centre for Mineral and Coal Technology (tekMIRA)

Jalan Jendenderal Sudirman No.623, 40211 Bandung

\*Corresponding e-mail: [slamet.handoko@esdm.go.id](mailto:slamet.handoko@esdm.go.id)

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| ABSTRACT  Government of Indonesia has made policies to increase the added value of coal. It can be done by using the technology of slow pyrolysis. Therefore, the aim of this study was to increase the added value of coal. This study was carried out by analysing the effect of temperature and water content of the feed to the products of slow pyrolysis. Water content variation in the feed form of coal is 10.57 wt% and 16.86 wt%. Temperatures variation in slow pyrolysis process were 500, 600, 700, and 800 ºC. The result of this research showed that low moisture content coal produced more char in slow pyrolysis process. Based on the variations of the temperature used, the higher temperature of pyrolysis produced fewer char but higher calorific value of coal product. The optimum operating condition was obtained at 500-600 ºC. |
| Keywords: slow pyrolysis, water content, temperature, char quality, and char energy content. |
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| *ABSTRAK*  *Pemerintah Indonesia telah membuat kebijakan untuk meningkatkan nilai tambah batubara. Hal ini dapat dilakukan dengan menggunakan teknologi pirolisis lambat. Oleh karena itu, tujuan dari 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 bentuk 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 menghasilkan lebih sedikit arang tetapi nilai kalor produk batubara lebih tinggi. Kondisi operasi optimum diperoleh pada 500-600 °C.* |
| *Kata kunci: pirolisis lemah, kandungan air, suhu, qualitas arang, dan kandungan energi arang.* |

Introduction

Based on data from the Geological Agency of the Ministry of Energy and Mineral Resources of Indonesia, 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 <5,100 kcal/kg of caloric content. Government of Indonesia has made regulations no 25 in 2018 on the Implementation of Mineral and Coal Mining Activities to increase added value for mineral and coal commodities.

Increasing the quality of coal can be done through 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 pyrolysis process is a thermochemical process in an *inert* or limited oxygen environment in which coal or biomass is slowly heated to a certain temperature and held for a set time in such a way as to produce complete degradation of the coal or biomass components to a solid product which has mass and energy gain. the maximum (Basu, 2013).

The suitable type of pyrolysis to be developed is slow pyrolysis because beside improving the quality of coal, a by-product can also be obtained in the form of coal tar which can be processed further into synthetic crude and coke oven gas(COG). Slow pyrolysisis pyrolysis which takes place rapidly at temperatures in the range 500 - 950 °C with a heating rate of around 10 - 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 pyrolysisprocess. When the ratio of hydrogen to carbon (H/C) decreases, the heating value of coal increases (Basu, 2013). Increasing the quality of coal will increase the price of coal.

The characteristics of the solid product from slow pyrolysis process(high quality coal) are influenced by the parameters of the slow pyrolysisprocess. Some of the parameters that affect the slow pyrolysisprocess 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 with the pyrolysis process by producing a low pyrolysis process temperature range between 200 - 300 °C (Bergman and Kiel, 2005; Tumuluru *et al.*, 2011; Rouset *et al.*, 2012; Sadak and Negi, 2009). The result of their coal pyrolysis research showed that the coal pyrolysis process was able to produce coal that has higher heating value than the feed.  The purpose of this study was to determine the effect of slow pyrolysistemperature and coal moisture content to coal product quality.

Experimental details

Equipment and Material

The equipment used in this study were coal screw feeder, fixed bed reactor, heat exchanger (HE)/condenser, and coal gas analyzer. Fixed bed reactor were 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 (N2), and anisol (benzene methoxy).



Figure 1. Test equipment configuration of pyrolysis process

Experiment Procedure

There were two variables in this study. The first variable was fixed variable condition: 20 gram/hour of coal and -32+42 mesh of coal size. The second variable was changed variable which consisted of reactor temperatures (500, 600, 700, and 800 ºC) and moisture of coal (10.57 wt% 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, pyrolysis process was carried out in the continuous scheme. A 20 grams of +32 mesh coal were put into the reactor for 1 hour and tested according to reactor temperature variation (500, 600, 700, and 800 ºC).

The experiment was started by flowing nitrogen gas into the pyrolysis equipment. Nitrogen gas (N2) was streamed to the reactor and gas cooling equipment to remove trapped air. The nitrogen gas flow rate was regulated at 24 mL/minute. Gas products flowed through the top of the reactor and then cooled through a condenser. Nitrogen gas was also used as a carrier gas during the experiment. When the experiment occurred, the nitrogen flow rate used was 0.05 mL/min. The gas out of the condenser was analyzed with a gas analyzer.

The pyrolysis reactor was set at 10 ºC/minute until temperature was reached. A 20 gram/hour of coal was fed into 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 routed to 4 pieces of impinger. The first 3 units of impinger were containing 60 mL of anisol and the last impinger was filled with activated carbon to ensure all tar were removed from NCG. The clean NCG was connected to a coal gas analyzer and gas composition analysis was performed. The composition of the NCG measured was CO, CO2, H2, CH4, O2, CnHm, and N2.

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. Proximate, ultimate, and calorific value of char were analyzed with ASTM method. Heat and material balance were obtained by calculation. A 100 kg of coal was used as the basis for the calculation. The energy content of coal was obtained from multiplication of 100 kg with calorific value of coal. The mass of char was obtained from multiplication of 100 kg with %char. Energy content of char was obtained from multiplication of mass of char with calorific value of char. While energy in char was obtained from division of energy content of char with 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 are 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 A have lower moisture content than Coal B. Therefore, at the same energy, less char was produced from Coal B than Coal A because the energy is used to evaporate water. This was also indicated by the more liquid product from Coal B than Coal A.

Char yield tend to flat from 500 ºC to 600 ºC then significant decrease in char yield occurred from 700 ºC 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 will require 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.

Figure 2. Material balance of slow pyrolysis

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 char produced will have lower 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 increased carbon and fixed ash.

The carbon content in char has increased after the pyrolysis process. 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. 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 was produced at high pyrolysis temperature contains low volatile matter and more carbonized carbon. Therefore, high carbon products can be 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). It 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).

Table 1. Properties of coal A

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Parameter | Unit | Coal A | Coal 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. Propertiesof coal B

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Parameter | Unit | Coal B | Coal 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 |

An increased in pyrolysis temperature between 500-700 ºC increased the calorific value of char sharply (Table 3 and 4). This was caused by the increased 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, K., Sharma, A.K., Lal, P.S., Dutta, S., and Maity, S., 2016).

Char Energy Content

The energy content of char to coal resulted from the calculation of mass and energy balance were showed in Table 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 can be utilized further.

Conclusions

Low moisture content coal produced more char in slow pyrolysis process. 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 need to be investigated further so the pyrolysis technology can be applied in an integrated scheme at the power plant.

Table 3. Coal A char analysis

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Parameter | Unit | Coal A | Coal 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 | Coal 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 |

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