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THE EFFECT OF HYDROTHERMAL PROCESS ON INCREASING HEATING VALUE AND REDUCING MOISTURE CONTENT OF LOW-RANK COAL

PENGARUH PROSES HIDROTERMAL TERHADAP PENINGKATAN NILAI KALOR DAN PENURUNAN KADAR AIR PADA BATUBARA PERINGKAT RENDAH

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ABSTRACT

As a low-rank coal, brown coal is characterized to have high water content. It is required a method to improve coal quality that can be used for effective coal utilization. One of the methods is to apply thermal dewatering technology which focuses on improving the quality of low-rank coal using water medium and autoclave. The purpose of this study was to determine the effect of coal to water ratio (w/v) and the effect of particle size of a low rank-coal on water content and calorific value using the hydrothermal method. Coal was processed under hydrothermal conditions at 150 °C for 60 min. For a mixture of 100 g of 25 mesh coal and 50 mL of water, the calorific value and water content after the hydrothermal process reached 5,849 kcal/kg and 5.10% (adb), respectively. While for a mixture of 100 g of 40 mesh coal and 50 mL of water, the calorific value and water content after the hydrothermal process reached 5,789 kcal/kg and 4.94% (adb), respectively. The calorific value obtained increased from the initial value of 3,296 kcal/kg and the water content decreased from the initial value of 44.34% for the coal condition before hydrothermal process. It can be concluded that the hydrothermal process with heating without oxygen carried out by a hydrothermal reactor could increase the calorific value of low-rank coal and reduce water content.

Keywords: hydrothermal; low-rank coal; calorific value; water content.

ABSTRAK

Batubara coklat sebagai batubara peringkat rendah mempunyai kandungan air yang tinggi. Diperlukan metode untuk meningkatkan kualitas batubara yang dapat digunakan untuk pemanfaatan batubara secara efektif. Salah satu caranya adalah dengan menerapkan teknologi hidrotermal yang fokus pada peningkatan kualitas batubara peringkat rendah dengan media air dan alat autoklaf. Tujuan penelitian ini adalah untuk mempelajari pengaruh dua variabel proses hidrotermal yaitu perbandingan batubara dengan air (b/v) dan ukuran partikel batubara peringkat rendah, terhadap kadar air dan nilai kalor. Pada proses hidrotermal, batubara dipanaskan pada suhu 150 °C dengan waktu pemanasan ±60 menit. Untuk campuran 100 g batubara berukuran 25 mesh dan 50 mL air, nilai kalor dan kadar air setelah proses hidrotermal masing-masing mencapai 5.849 kkal/kg dan 5,10%. Sedangkan untuk campuran 100 g batubara berukuran 40 mesh dan 50 mL air, nilai kalor dan kadar air mencapai 5.789 kkal/kg dan 4,94%. Nilai kalor yang diperoleh ini meningkat dari nilai awal sebesar 3.296 kkal/kg dan kadar air menurun dari nilai awal sebesar 44,34% untuk kondisi batubara sebelum mengalami proses hidrotermal. Dapat disimpulkan bahwa proses hidrotermal dengan kondisi dan variabel proses penelitian dapat meningkatkan nilai kalor batubara peringkat rendah dan menurunkan kadar air.

Kata kunci: hidrotermal; batubara peringkat rendah; nilai kalori; kadar air.

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INTRODUCTION

As reported in the Handbook of Energy & Economic Statistics of Indonesia 2023, Indonesia has total coal resources of 97,297.11 million tons with verified coal reserves of around 30,218.98 million tons. Around 62% of the reserves are lignite or low-rank coals (Arinaldo and Adiatma, 2019). Low rank coals generally have a high water content of around 40% by weight and a low calorific value. Their use is still limited in industry. It is known that coal production in 2023 is estimated to reach around 775.2 million tons, with total exports of 518.05 million tons (Adi, 2024). According to D. Zhang *et al.* (2016). Low-rank coals have high water content of 25-60%. Katalambula and Gupta (2009) also explained that coal is categorized as low-rank coal if it has one or more problematic properties related to its use in power plants, including (i) low calorific value and high water content, which may cause low efficiency, (ii) low volatile content, which affects flame stability, (iii) high ash content, which is related to ash and efficiency problems. This coal requires special technology for its application, especially in power plants. According to Katalambula and Gupta (2009), this problem requires special technology to solve it, including mechanical thermal expression and hydrothermal dewatering. Both are non-evaporative drying/dewatering technologies.

Treatment can be carried out through a hydrothermal drying process to reduce water content and increase calorific value. Hydrothermal processes require smaller reactors that can reduce investment costs (Huda, Salinita and Ningrum, 2017). In steam power plants (PLTU) and industry, low-rank coals cause various problems related to efficiency and operational costs. Low-rank coals have a higher moisture content. This causes incomplete combustion, because some of the energy from the coal is used to evaporate water rather than to generate electricity. As a result, combustion efficiency decreases and produces more emissions. Low-rank coals often contain minerals and other impurities that can form scale or deposits inside the boiler. This scale can insulate the heating surface, reduce heat transfer, and cause a decrease in the efficiency of the heat exchanger. In the long term, this can shorten the life of the equipment and increase maintenance costs. In addition, high moisture and ash content cause high transportation costs.

The efforts made to improve the quality of low-rank coals are by reducing the water content and increasing the calorific value by heating the coal using the hydrothermal method (Ifa *et al.*, 2019). Hydrothermal technology in low-rank coal aims to reduce the water content in coal. The process of drying coal at a certain temperature can change the chemical properties of coal, which will affect the effectiveness of the conversion process. The hydrothermal drying process of coal at temperatures above 150 °C had caused changes in the oxygen functional groups and produce coal having higher carbon and lower oxygen contents (Huda, Salinita and Ningrum, 2017).

Several studies on hydrothermal dewatering processes have been conducted in the range of 150-300 °C. Research conducted by Murray and Evans (1972) showed how heating coal in the pressurized water could affect the water content. At temperatures between 250 and 300 °C, three-quarters of the original water content of the coal could be removed. This shows that high temperatures could effectively increase the removal of water from coal. After heating, when the coal was cooled in water at 150°C, water reabsorption into the coal structure occurred completely. This may be due to the porosity of the coal left after water removal, which allows water molecules to be re-adsorbed. However, when the treatment temperature was increased, water reabsorption decreased. Yu *et al.* (2012) explained that the hydrothermal dewatering method at temperatures of 150 to 380 °C for peat (Mursito *et al.*, 2010; Mursito, Hirajima and Sasaki, 2010), biomass (Sun *et al.*, 2011), and other low-quality coal (Mursito, Hirajima and Sasaki, 2011) could improve the quality of the material including removing water, sulfur, and ash content, as well as improving the physical and chemical properties of the raw materials. Y. Zhang *et al.* (2016) reported that the hydrothermal dewatering of Shengli lignite from China was carried out at temperatures of 200–300 °C. This process showed a significant reduction in the equilibrium moisture content (M_{eq}) of lignite. At a temperature of 200 °C, the equilibrium moisture content decreased from 29.75% in the raw coal to 13.59%. While at a temperature of 300 °C, the equilibrium moisture content further decreased to 7.40%. This decrease was indicated by the decomposition of oxygen functional groups and changes in the hydrophilic properties of the coal samples. The results of this study illustrate the effectiveness of hydrothermal drying in improving the quality of lignite by reducing the moisture content.

Hydrothermal treatment of lignite carried out at different temperatures (200, 250, 300, and 350 °C) by Wan *et al.* (2019) showed that the process had a significant impact on the physical and chemical properties of lignite. Through this treatment, the removal of oxygen-containing functional groups could help improve the stability and quality of lignite, as well as reduce the ability of lignite to retain water. According to Liu *et al.* (2021), lignite from Inner Mongolia Baiyinhua No. 2 mine was used as raw material with particle size less than 0.5 mm, which was processed at various temperatures of 150, 200, 250, and 310 °C. The different temperature treatments were aimed to observe the changes in physical and chemical properties of lignite due to the loss of oxygen-containing functional groups. The results showed that the hydrothermal dewatering process not only improved the coal rank but also increased the hydrophobicity of lignite, as well as reduced its surface hydrophilicity. This is important for coal applications, especially in the context of energy generation and coal conversion processes.

In this study, the effect of varying lignite particle sizes of 25, 40, 60, and 120 mesh and coal to water ratio (w/vol.) at 150 °C in hydrothermal process on the calorific value and water content were investigated. The increase in calorific value and decrease in water content will be visible from the graphic profile obtained.

METHODOLOGY

Sampling in the Field

The coal samples used in this research were low-rank coal from PT. Bhumi Sriwijaya Perdana Coal, Musi Banyuasin Regency, South Sumatra. Sampling in the field was carried out using a channel sampling procedure as shown in Figure 1. This sampling was carried out by taking samples on the coal wall, taking each part of the layer at a certain distance and depth, as shown in Figure 2. Figure 1 shows the length of the entire sample. The coal in the field that will be taken for analysis was 2.60 meters long and was divided into five to take coal samples from each layer, where four plastic bags of samples were taken from each layer of coal, total samples of 20 plastic bags from the five layers of coal that have been divided previously.

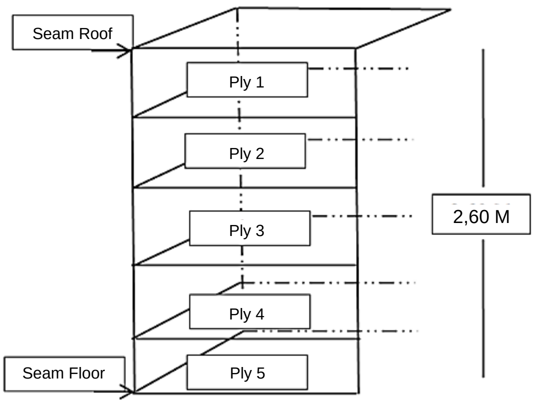

Figure 1. Sampling scheme in the field

a. b.

Figure 2. Sampling stages: (a). Measurement of the length of coal samples in the field, (b). Coal sampling in the field

Preparation of Coal

Coal samples were reduced in size with a crusher. The crushed coals were sifted into 100 g with sizes of 25, 40, 60, and 120 mesh. Before undergoing the hydrothermal process stages, coal samples were tested for calorific value and water content.

Hydrothermal Process

Hydrothermal process was carried out in the reactor shown in Figure 3. A hundred grams of coal with varying sizes in a mesh was added to a certain volume of water while stirring for 3 min. Then, this mixture was fed into a hydrothermal reactor to undergo the slurry dewatering process at 150 ºC for 60 min and cooled to room temperature for ±60 min. Coal and water were separated through a drying process in an oven because the mixture was in the form of a slurry (the amount of solid was more dominant than the liquid). Although according to Y. Zhang *et al.* (2016), the separation of the mixture of solids and liquids was through filtration because this study used a composition of coal and water (100 g: 250 mL), so water could be separated through a filtration process as much as 2 times. Then, coal samples were tested for water content and calorific value after the upgrading process.

Figure 3. Hydrothermal Process

RESULTS AND DISCUSSION

Table 1 shows the quality of raw and upgraded coals. According to ASTM D388, raw coal is classified as lignite B with a calorific value of 6,043 BTU/lb (mmmf). After undergoing the upgrading process, the calorific value of the coal increased to 10,640 BTU/lb (mmmf). It can be said that the resulted coal from the upgrading process is classified as Subbituminous A Coal.

Effect of Coal to Water Ratio on Calorific Value and Water Content

Figure 4 shows that after the hydrothermal process, there was an increase in the calorific value from the initial calorific value of 3,296 kcal/kg to 5,849 kcal/kg with adding water 50 mL. This increase occurred because

Table 1. Coal analysis of raw and upgraded coals (air-dried basis)

Analysis Parameters	Coal samples		Standard Methods
Proximate (adb, %)	Raw Coal	Upgraded Coal*	
Moisture (M)	44.34	4.42	ASTM D.3173
Ash (A)	1.78	2.86	ASTM D.3174
Volatile Matter (VM)	31.58	45.94	ASTM D.3175
Fixed Carbon (FC)	22.30	46.78	ASTM D.3172
Ultimate (adb, %)			
Total Sulfur (TS)	0.37	0.52	ASTM D.4239
Carbon (C)	36.14	62.30	ASTM D.5373
Hydrogen (H)	7.16	4.70	ASTM D.5373
Nitrogen (N)	0.70	1.20	ASTM D.5373
Oksigen (O)	53.85	28.42	ASTM D.3176
Gross Calorific value (kcal/kg, adb)	3,296	5,726	ASTM D.5865
Calorific value** (Btu/lb, mmmf)	6,043	10,640	ASTM D.388

* At a temperature of 150 ºC for 60 min on a mixture of 25 mesh coal and 50 mL water

** $CV\ (mmmf) = 100 \times \frac{CV\ adb - (50 \times TS)}{(100 - (1.08 \times A + 0.55 \times TS))}$

CV = calorific value

(Azizi and Ghifari, 2020)

of the effect of using fewer water media compared to other samples, so that the influence of temperature, pressure, and long heating time of ±1 h caused the water in the coal to evaporate faster than other coal samples that used more water. The results of this study are supported by the results reported by Murray and Evans (1972), the phenolic group has begun to break down at a temperature of 150 ^oC. The main product of this group decomposition is most likely water. Liu *et al.* (2016) reported that the hydrothermal process is a process to increase the calorific value of coal by reducing water content. According to Liu *et al.* (2022), hydrothermal conditions allow the removal of oxygen-containing functional groups, which cause changes in surface characteristics. This happens because the functional group containing oxygen very easily forms hydrogen bonds with water, resulting in stronger hydrophilicity and high water content. This is also following what was reported by Ge *et al.* (2018). When the hydrothermal dewatering temperature was below 250 °C, the destruction of active groups, such as methylene, methoxyl, and carboxyl groups, induced the formation of micropores and the expansion of the pore structure. It could be said that the pore structure developed, the surface area and total volume increased. Wu *et al.* (2015) reported that high temperatures and pressures caused the release of free water, built-in water, tar, hydrogen, CO_2, CO, and hydrocarbons. Tar coming out of the coal might cover the pores of the coal surface that are open due to the heating process, so that the water coming out could not be reabsorbed by the coal. According to Ge *et al.* (2015, 2018), this phenomenon occurred through the hydrothermal dewatering process of coal at a temperature of 250-300 °C.

A quite sharp decrease in calorific value occurred when 60 and 70 mL of water were added to the coal. However, overall, based on the trend of the calorific value profile due to increasing the amount of water, it shows that the calorific value obtained is still in the range of 5,729 - 5,849 kcal/kg. When water was added to coal, the water content could replace some of the solid fuel, thereby reducing the concentration of carbon, which is the main component that produces energy during combustion. In this regard, van Raam, Ruyter and van Breugel (1981) also carried out coal dewatering and upgrading at a temperature of 150-300 °C at a pressure that prevented water evaporation.

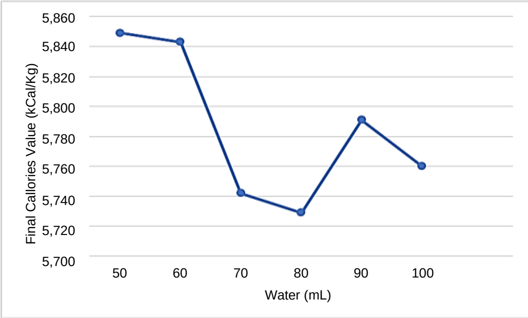

Figure 4. Effect of Coal to Water Ratio (w/v) on the Calorific Value (Coal of 25 mesh = 100 g, T = 150 ^oC)

Figure 5 shows the total water content in the coal sample has decreased after hydrothermal analysis. The lowest water content after the upgrading process reached 5.1% from 44.34% with the addition of 50 mL of water.

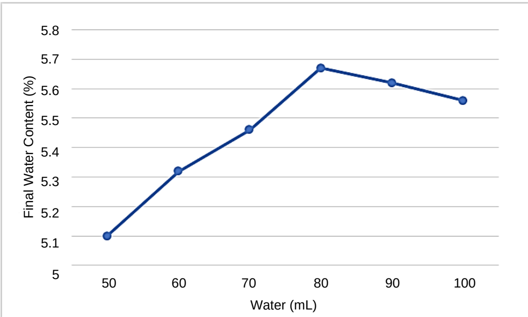

Figure 5. Effect of Coal to Water Ratio (w/v) on the water content (Coal of 25 mesh = 100 g, T = 150 ^oC)

Effect of the particle size of coal on calorific value and water content

Figure 6 shows that there was an increase in the calorific value after coal was upgraded from 3,296 to 5,789 kcal/kg for a coal particle size of 40 mesh. When the particle size was reduced from 40 mesh (0.420 mm) to 120 mesh (0.125 mm), the heating value decreased slightly, but the values were still relatively high, namely 5,726 and 5,730 kcal/kg.

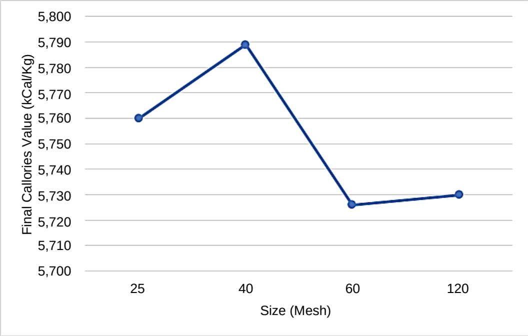

Figure 6. Effect of the Particle Size of Coal on the Calorific Value (Coal = 100 g, water = 100 mL, T = 150 ºC)

Figure 7 shows that there was a decrease in the water content of coal as the particle size became smaller. When the hydrothermal process of 60 mesh coal with 100 mL water added was carried out, the water content decreased from 44.34 to 4.42%. Overall, for all the particle sizes, the water content decreased by an average of about 87%. Water content decreased along with the smaller particle size. As the particle size of coal decreased, the surface area of the particles increased, which could increase the reactivity of coal. This was because during processing, the oxygen-containing functional groups in coal would be decomposed, as reported by Liu *et al.* (2021) and Sa'ban *et al.* (2022). This is why the smaller the particle size, the easier the tendency of thermal reactions is to reduce the oxygen content, which consequently can contribute to the changes in the thermal and chemical properties of coal.

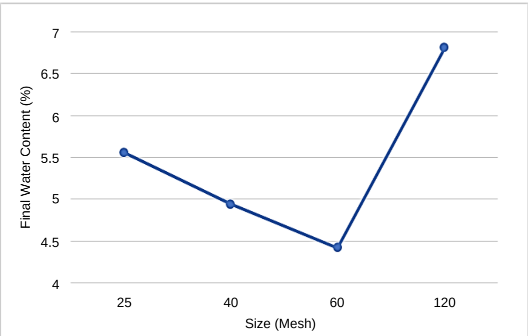

Figure 7. Effect of the Particle Size of 100 g Coal on the Water Content. (Coal = 100 g, water = 100 mL, T = 150 C)

As investigated by Liu *et al.* (2021), the particle size of the coal to be upgraded helped the process of reducing water content during the hydrothermal dewatering process, which was indicated by the reduction of groups containing oxygen atoms. The particle size of the lignite used was less than 0.5 mm (35 mesh), which indicated that during the hydrothermal dewatering process at 150 °C there has been a removal of groups containing oxygen atoms.

Murray and Evans (1972) explained the phenomenon of moisture content removal on thermal dewatering of brown coal in water systems. The functional group analyzes are used to explain the removal of liquid water. The phenolic group begins to decompose at a temperature of 150 °C, and at temperatures above 200 °C, the damage is quite obvious. The main product of decomposition of this group is probably water. Free carboxylic acid groups are stable up to 150 °C and carboxylate groups decompose from 20-200 °C in small amounts.

CONCLUSIONS

Hydrothermal is an effective coal upgrading process that can improve coal ranking based on calorific value and water content. After undergoing the upgrading process, the calorific value of the coal increased to 10,640 BTU/lb (mmmf) and moisture content decreased to 4.42% (adb). It can be said that the resulted coal from the upgrading process was classified as Subbituminous A Coal. This value was obtained when 100 g of coal (20 mesh) was hydrothermally heated in 50 mL of water at 150 °C for 1 h. Before upgrading, the calorific value of coal was 5,726 BTU/lb (mmmf) or 3,296 kcal/kg (adb) and the water content was 44.34% (adb).

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