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# HEAVY OIL FROM CATALYTIC CRACKING OF POLYPROPYLENE-LOW DENSITY POLYETHYLENE PLASTIC WASTE PYROLYSIS OIL TO IMPROVE THE QUALITY OF LIGNITE

## *PEMANFAATAN MINYAK BERAT HASIL PERENGKAHAN KATALITIK MINYAK PLASTIK* POLYPROPYLENE-LOW DENSITY POLYETHYLENE *UNTUK MENINGKATKAN KUALITAS LIGNIT*

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**ABSTRACT**

The most obvious characteristics of lignite are its high moisture content (30-50%) and relatively low calorific value compared to other types of coal. This causes low combustion efficiency, making it not optimal for use as fuel. In this study, the slurry dewatering process was applied to coal that has a low calorific value of 3,662 cal/g and a moisture content of 37.29%. The addition effect of heavy oil from catalytic cracking of Polypropylene-Low Density Polyethylene (PP/LDPE) plastic waste pyrolysis oil on improving quality of lignite was investigated in this study. Heavy oil was used as additive. The ratio of 120 mesh lignite to heavy oil used was 100:15, 100:30, 100:45, 100:60, 100:75, and 100:90 (g/mL). Coal was carried out with a slurry dewatering process in an autoclave at various temperatures of 140 and 150 °C for 60 min. The test results obtained the highest calorific value of 6,374 cal/g and the moisture content of 2.81% for a ratio of lignite to heavy oil of 100: 45 g/mL at a temperature of 140 °C.

Keywords: lignite; heavy oil; PP/LDPE; slurry dewatering; moisture; calorific value.

***ABSTRAK***

*Karakteristik lignit yang paling jelas adalah kadar airnya yang tinggi (30-50%) dan nilai kalor yang relatif rendah dibandingkan dengan jenis batubara lainnya. Hal ini menyebabkan efisiensi pembakarannya rendah, sehingga kurang optimal untuk digunakan sebagai bahan bakar. Dalam penelitian ini, proses dewatering slurry diterapkan pada batubara yang memiliki nilai kalor rendah yaitu 3.662 kal/g dan kadar air 37,29%. Pengaruh penambahan minyak berat hasil perengkahan katalitik minyak pirolisis limbah plastik Polypropylene-Low Density Polyethylene (PP/LDPE) terhadap peningkatan kualitas lignit dipelajari dalam penelitian ini. Minyak berat digunakan sebagai zat aditif. Perbandingan lignit 120 mesh dan minyak berat yang digunakan adalah 100:15, 100:30, 100:45, 100:60, 100:75, dan 100:90 (g/mL). Batubara diproses dengan metode* slurry dewatering *dalam autoklaf pada suhu 140 dan 150 °C selama 60 menit. Hasil pengujian diperoleh nilai kalor tertinggi sebesar 6.374 kal/g dan kadar air 2,81% untuk perbandingan lignit terhadap minyak berat 100:45 g/mL pada suhu 140 °C.*

*Kata kunci: lignit, minyak berat,* PP/LDPE, slurry dewatering, moisture, *nilai kalori*.

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## INTRODUCTION

Coal is one of the most important materials in national development. Along with the increase in oil and gas prices, coal becomes an alternative that is expected to overcome energy shortages to increase its utilization in domestic purposes such as power generation fuels, and so on (Kadir, Widodo and Anshariah, 2016). According to the Ministry of Energy and Mineral Resources of the Republic of Indonesia, in 2020, there is abundant coal in Indonesia, especially in Sumatra which reached 64,592.37 million tons. The total coal reserves of 14,799.99 million tons have varying quality, ranging from high-calorie to low-calorie coal. Mining efforts have been widely undertaken in some places against low-calorie coal on both a large and small scale. With characteristic coal on the island of Sumatra, which has a reserve of lignite about 60% (Heriyanti *et al.*, 2019).

Lignite has a high moisture content on average between 30-50% but has a low ash and sulfur content (Tua, Wibowo and Rosyid, 2020). Calorific value ranging from 868 to 2,196 kcal/kg are considered low quality, whereas values between 2,200 and 5,574 kcal/kg are deemed high quality (Aytaç Korkmaz, 2020). According to Umar, Monika and Suganal (2020), the use of low-calorie coal can affect the environment, with high sulfur reservoirs that are very harmful to the environment. Sulfur content can cause acid rain while the content of flying ash is very dangerous for human health, in addition, high levels of ash and sulfur can increase the company's operating costs. For optimal utilization of low-calorie coal, the help of technology is needed to increase the value of low-grade coal calories so that it becomes a mainstay commodity.

The use of technology will increase the economic value of initially less useful low-calorie coal to be more beneficial (Heriyanto *et al.*, 2014; Afrah *et al.*, 2023). One of the efforts to increase the economic value of lignite is the use of up-grading technology. There are several coal up-grading processes developed today, including using the principle of evaporation, namely, the Steam Tube Dryer (STD), Steam Fluidized Bed Dryer (SFBD), and Slurry Dewatering (UBC) methods. Non-evaporation is a method of mechanical dewatering, Fleissner process and Hot Water Dewatering (HWD).The principles of pyrolysis include the Encoal and K-Fuel (Koppelman process) methods. These processes are distinguished based on the reaction conditions, type of heat transfer media, type of reactor, quality of liquid waste, and processing costs.

Applications of low-ranking coal up-grading process technology should consider several important aspects that are techno-economically feasible and to a minimum involving chemical changes in coal. This means that the up-grading process at low temperatures and pressures is more desirable so that the processes of releasing organic compounds or pyrothic processes can be suppressed so that liquid waste and gas emissions produced are almost non-existent. Operating at low temperatures and pressures will also lower operating costs in a significant amount (Hartiniati, 2007).

The technology proposed in this study is slurry dewatering, which is a technology to increase the calorific value of lignite with relatively simpler operating conditions, with a temperature of 140-150°C and pressure of 2-3 atm (Heriyanto *et al.*, 2014). Shigehisa, Inoue and Kumagai (2015), successfully raised the calorific value of coal with the upgrading method from 3,500 to 6,000 cal/g by lowering the moisture content and coating the surface of coal with coating.

In this study, an increase in the calorific value of low-quality coal with the slurry dewatering method using the addition of heavy oil produced from catalytic cracking of Polypropylene-Low Density Polyethylene plastic waste pyrolysis oil. This heavy oil is used as an additive for lignite because it has some chemical properties in common for coal, with some similarities this property makes this residual oil able to enter coal pores. This agrees with the research of Jaya *et al.* (2017).The heavy oil is a liquid containing naphtha and other components that have relatively potential to be reprocessed into fractions or something that can provide added value. With the application of heavy oil for the process of upgrading lignite is very appropriate to do (Philp, 2020). Based on the heat value of plastic materials, Polypropylene-Low Density Polyethylene (PP/LDPE) plastic type has a large heat value of 46.4 MJ/kg compared to other types of plastic materials. The chemical properties of polypropylene have excellent resistance to non-oxidizing inorganic chemicals, detergents, alcohols and so on. However, polypropylene can be degraded by oxidizing agents such as nitric acid and hydrogen peroxide. Its high crystallinity properties cause its strain power to be high or stiff and hard (Panda, Singh and Mishra, 2010; Nugroho, Alhikami and Wang, 2023).

Some research about increased quality coal has been done previously by some researchers. Billah (2010) has done research on the increased quality of low-rank coal with used soil oil and residue oil through process slurry dewatering. Putri and Fadillah (2020) has conducted research on increasing the caloric value of low-ranking coal by using UBC process technology with using residue oil from lubricant former to improve the quality of coal PT. Cahaya Bumi Perdana in West Sumatra Province. Billah (2010) showed that calorific value from low-rank coal with a rating from 4,702 to 6,692 kcal/kg, moisture content of 0.668%, ash content of 11.883%, volatile matter of 30.122%, and fixed carbon of 57.377%. Putri and Fadillah (2020) reported that the upgrading process can increase the calorific value of coal from 3,177.76 to 7,122.26 kcal/kg. The results of their proximate analysis were moisture of 1.17%, volatile matter of 62.73%, and ash of 22.55%, with the ratio of used lubricating oil and coal was 1:4, 1:2 and 3:4.

Therefore, researchers chose the slurry dewatering process as the process of upgrading lignite, with heavy oil from catalytic cracking of Polypropylene-Low Density Polyethylene plastic waste pyrolysis oil as an additive to improve the quality of lignite. It is expected that the use of heavy oil can increase the calorific value of lignite.

## METHODOLOGY

### Material and Method

This study used lignite from PT. Bhumi Sriwijaya Coal, South Sumatra with a calorific value of 3,662 cal/g and a water content of 37.29%, as shown in Table 1. The additive used in this study was residual oil from the catalytic cracking of mixed PP/LDPE plastic oil. This residual oil is brownish in color and has a calorific value of 11,000 kcal/kg (ASTM D3173/D3173M-17) and a moisture content of 0.024%. Both materials are shown in Figure 1.

Table 1. Characteristics of Raw Coal

| Parameters | Result (adb) | Method |
|---|---|---|
| Inherent Moisture (%) | 37.29 | ASTM D 317M-17 |
| Ash Content (%) | 4.27 | ASTM D 3174-12 |
| Volatile Matter (%) | 33.04 | ISO 562:2010 |
| Fixed Carbon (%) | 25.41 | ASTM D 3172-12 |
| Gross Calorific Value (cal/g) | 3,662 | ASTM D 5865M-19 |

Adb: air dried basis

a. b.

Figure 1. Lignite (a) and heavy oil from catalytic cracking of Polypropylene-Low Density Polyethylene plastic waste pyrolysis oil (b)

Lignite coal was ground with a crusher to obtain 120 mesh coal. The initial coal sample was first tested for calorific value and proximate analysis. Furthermore, 100 g of coal with a size of 120 mesh was dried at a temperature of 105°C for 4 hours so that the water content on the coal surface was reduced. Preparation of additives was carried out with the desired volume with variations of 15, 30, 45, 60, 75, and 90 mL. Dry coal powder was mixed with additives to form a slurry and then the slurry dewatering process was carried out at 140 and 150°C for 60 minutes. After this process was completed, treated coal was obtained as shown in Figure 2. Furthermore, the coal fraction and residual oil were separated using a vacuum filter. This coal was dried again at a temperature of 110°C for 60 minutes until a constant mass was obtained. The stages of the research process can also be seen in Figure 3. Coal that had been treated with additives was tested for moisture content, calorific value, FTIR analysis and SEM-EDX analysis.

Figure 2. Treated coal in slurry dewatering process

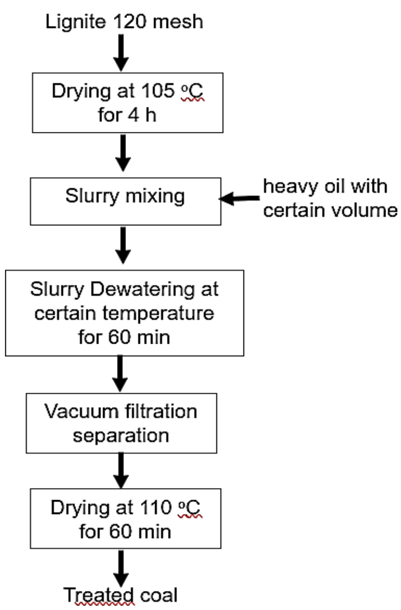



Figure 3. Slurry dewatering process

### Chemical Structure Analysis

The coal slurry dewatering test was carried out using an FTIR spectrometer (Nicolet Avatar 360IR). A total of 1 mg of coal sample after processing was mixed with KBr with a ratio of coal and KBr of 1: 200. The sample was pressed using a hydraulic press with a pressure of 7 torr. The spectrum results were measured at a wavelength of 4000-499 $cm^{-1}$ and analyzed by the curve fitting method.

### Physical Structure Analysis

The surface morphology of coal was tested using Phenom Dextom ProXL as shown in the SEM image with an accelerating voltage of 15 kV. The coal sample was prepared in the form of a layer with Au covered on its surface to increase the resolution. Similarly, EDX testing was performed on the same sample. The SE detector was placed on the device to detect the radiation energy. The sample in space was brought close to the electron source which was then emitted by the electron beam and the results were obtained. Then the analysis results were scanned and the percentage of elements contained in the coal was obtained.

## RESULT AND DISCUSSION

### Preliminary Coal Analysis

Coal was subjected to preliminary analysis in the form of proximate analysis and initial coal calorific value using ASTM and ISO methods. The results obtained are shown in Table 1.

Parameters that are used as references in determining coal quality include calorific value, moisture content, volatile matter, ash content, and Hardgrove Grindability Index or HGI (Widodo and Sulistyo, 2014). Based on the ASTM D388 classification, the characteristics of coal are included in the lignite category. This type of coal has a low selling value, because its calorific value is low, its moisture content is high and its carbon content remains low, so that if it is burned it will pollute the environment (Brotowati and Sofia, 2018).

### Effect of the ratio of coal to additives on moisture content and calorific value at temperature of 140ºC

After the slurry dewatering process, the moisture content in the coal has been reduced. The reduction in coal mass occured from the initial mass of 100 g. A comparison of moisture content between the initial and the final coal is shown in Figure 4.

The moisture content in the initial coal sample was 37.29%. Based on Figure 4, the lowest moisture content was found in dewatered coal (X6) of 1.30% with a ratio of 100: 90 (g/mL). The highest moisture content was found in dewatered coal (X4) of 3.20% with a ratio of 100: 60 (g/mL). The average decrease in moisture content in dewatered coal was

93.40%. This showed that the addition of additives with a ratio of 100:90 (g/mL) successfully diffused and coated the open coal pores. Based on reported by Umar (2010) who stated that the addition of additives in the slurry dewatering process could coated coal pores and reduce moisture content. According to Amarullah, one of the factors that affects the quality of coal is water content (Amarullah, 2007). Moisture content and fixed carbon greatly determine the calorific value of coal. Coal with low water content usually has better quality and higher calorific value compared to coal with higher moisture content. This is because high moisture content in coal will reduce combustion efficiency and produce less energy (Rahim and B.Z, 2012).

Based on Figure 5, the highest calorific value was obtained in coal from the dewatering process (X3) with a ratio of 100: 45 (g/mL) of 6,374 kcal/kg, indicating an increase in calorific value of 74.06%. The lowest calorific value was obtained in coal from dewatering (X1) with a ratio of 100: 15 (g/mL) of 5,821 kcal/kg, indicating an increase in calorific value of only 58.96%. It can be said that the slurry dewatering process affects the calorific value of the dewatered coal produced. Calorific value is one of the main determinants of coal quality, the calorific value produced from coal combustion depends on the high and low calorific value (Setiawan, Triantoro and Annisa, 2018).

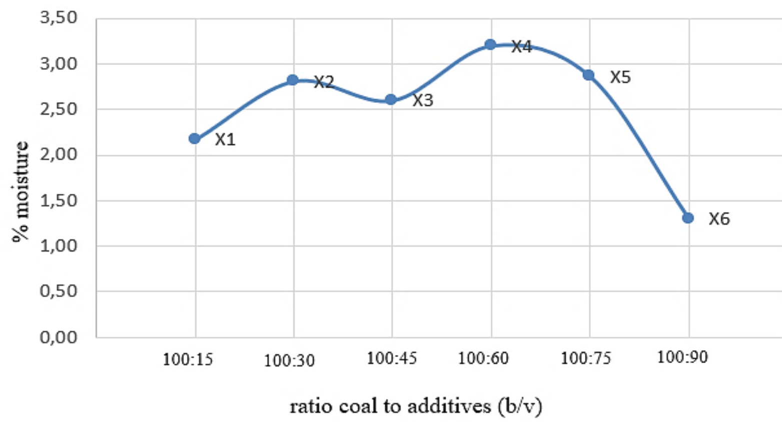



Figure 4. Effect of ratio coal to additives on the moisture content of dewatered coal at 140 ºC

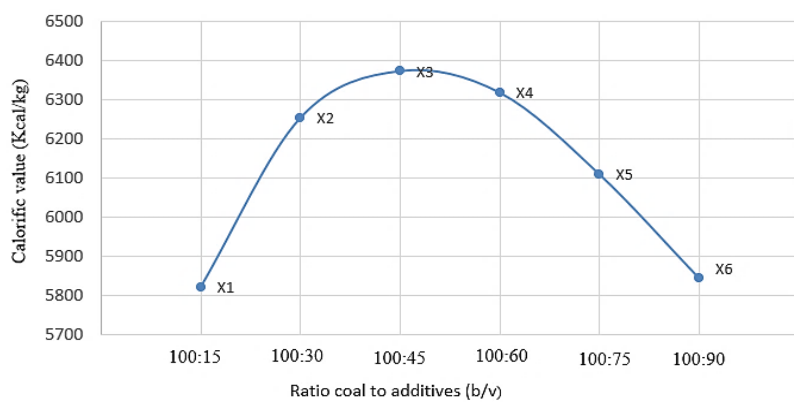



Figure 5. Effect of ratio coal to additives on calorific value of dewatered coal vs ASTM Coal Classification at 140 ºC

**Effect of the ratio of coal to additives on moisture content and calorific value at temperature of 150ºC**

It is known that the initial moisture content of the coal sample is 37.29%. Figure 6 shows that the lowest water content of 0.73% was identified in the dewatered coal (Z6) with a ratio of 100: 90 (g/mL). While the highest moisture content was obtained at 4.02% with a ratio of 100: 45 (g/mL). The average decrease in moisture content in dewatered coal was 93.08%.

The moisture content in dewatering coal at a temperature of 150 ºC tends to show an upward trend from Z1 to Z3 and then shows a downward trend from Z3 to Z6. The increase in moisture content from Z1 to Z3 occured due to low adsorption of additives, while the decrease in moisture content from Z3 to Z6 were due to adsorption of additives with a larger portion. According to Choi *et al.* (2011), the more additives were added, the moisture content decreased. Meanwhile, according to research conducted by Kadir, Widodo and Anshariah (2016), if the value of coal moisture content is low then the calorific value is high and vice versa. This deviation can be caused by the use of light oil fractions as additives, the accuracy of the tool temperature is still not optimal and the briquetting of the final sample is not carried out. So that the moisture stability of dewatered coal is not maintained (Malaidji, Anshariah and Budiman, 2018).

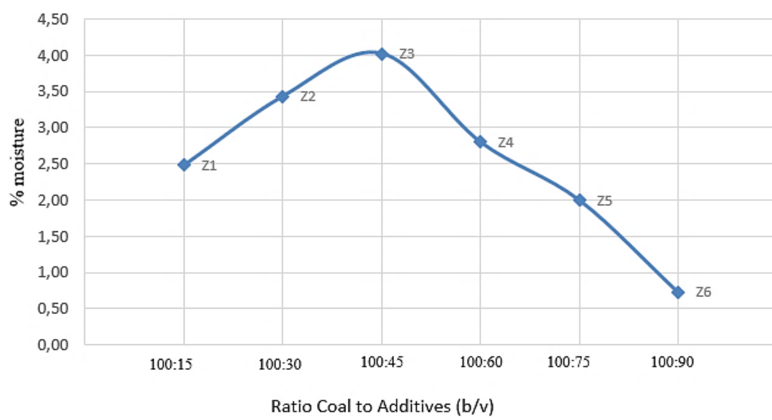



Figure 6. Effect of ratio coal to additives on the moisture content of dewatered coal at 150º C

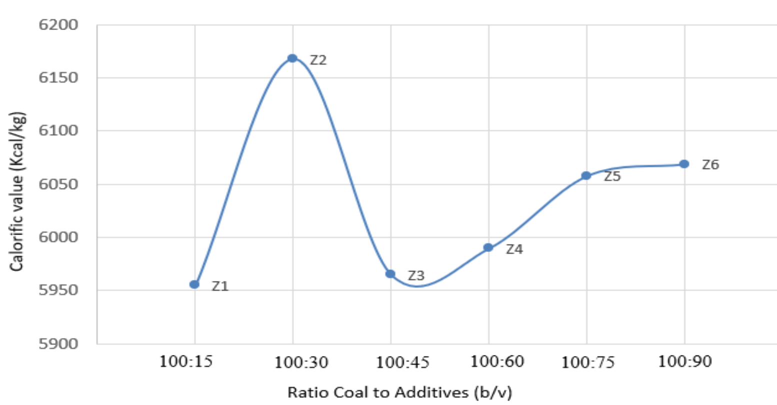



Figure. 7. Effect of ratio coal to additives on calorific value of dewatered coal vs ASTM Coal Classification at 150 ºC

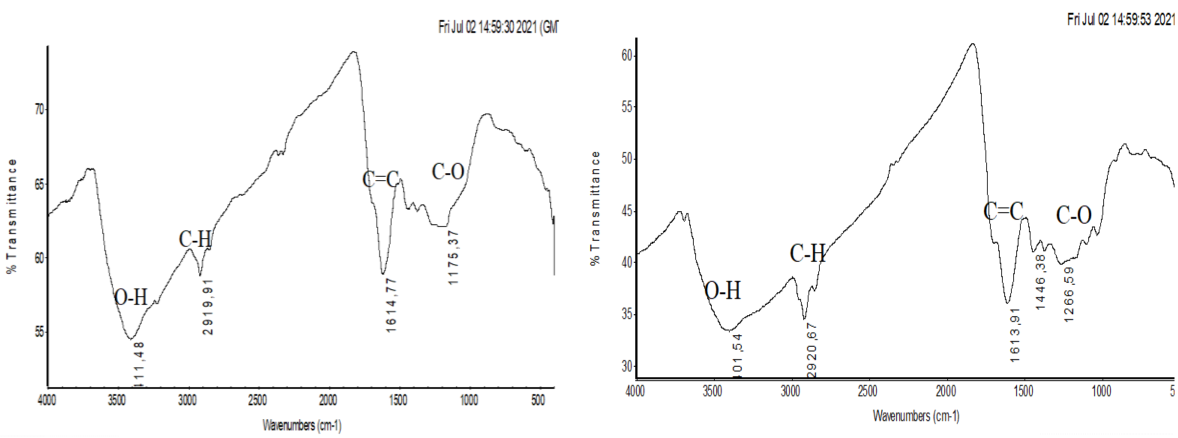
The dewatering slurry process has an influence on the calorific value of the dewatered coal produced. As shown in Figure 7, there is the highest calorific value of 6,168 kcal/kg in the Z2 coal sample with a ratio of 100: 30 (g/mL).The lowest calorific value was found in the Z1 coal sample of 5,955 kcal/kg with a ratio of 100: 15 (g/mL). At a ratio of 100: 30 (g/mL) was the optimum operating condition at this temperature. According to Jaya *et al.* (2017) if a ratio of coal to oil was greater with the size of the oil molecules being smaller than the size of the coal pores (macropores), this caused more heavy oil to enter the coal pores so that the calorific value became high.

**FT-IR Analysis**

FT-IR analysis is used to determine the functional groups in coal and their effects on coal characteristics such as oxygen functional groups. According to Feng *et al.* (2018) the water content in coal is related to the abundant oxygen content in functional groups (Feng *et al.*, 2018). Its presence affects the coal rank with the number and type of groups used to relate the coal rank characteristics.

Although Figure 8 shows that the trend of FTIR spectra of raw and upgraded coal are similar to each other, there has been a reduction in the intensity of the peaks. As reported by Murray and Evans (1972) that at a temperature of 150 $^oC$ the phenolic group started to break down. Sa'ban *et al.* (2022), Ge *et al.* (2015) and Feng *et al.* (2016) also found that the FTIR spectra of upgraded coal at 150, 250 and 300 $^oC$ were similar to raw coal.

The moisture content in coal is evidenced by the presence of hydroxyl groups (O-H). Hydroxyl groups have wave uptake around 3,700-3,100 $cm^{-1}$ (Skoog, Holler and Crouch, 2017). In the coal before treated, the presence of the O-H group was seen at wavelengths of 3,411.48 $cm^{-1}$ with an intensity of 54.38 %T. Meanwhile, in the final sample, the O-H group was seen in a wavelength area of 3,401.54 $cm^{-1}$ and had an intensity of 33.212 %T. The O-H group contained in the sample did not include the free group but had an O-H bond as evidenced by a widened uptake band (for O-H bonds) instead of a pointy or narrowed (free cluster) (Anam, Sirojudin and Firdausi, 2007). Based on the data, there was a loss of the O-H function group in the final coal. This indicated that there was a reduction in moisture content in the coal after treated. It could be said that the additives used successfully cover the pores of coal to push the moisture content contained in coal out and reduce the content of coal moisture.




a. b.

Figure 8. FTIR spectra of (1) coal before treated and (b) coal after treated

The main content of hydrocarbons (C and H) in coal were strengthened by the presence of C-H groups. It is known that the C-H group has an absorption area with a wavelength of 2,970-2,850 $cm^{-1}$ (Skoog, Holler and Crouch, 2017). Based on the results of the test on the initial sample and the final sample, the C-H group read in the absorption area of 2,919.91 and 2,920.67 $cm^{-1}$ which were classified as strong intensity. The content of C-H groups in early coal was classified as C-H alkane as evidenced by the absence of the appearance of C-H group around 1,500-1,300 $cm^{-1}$. This proved the increased calorific value of lignite (Nurhadi, 2017).

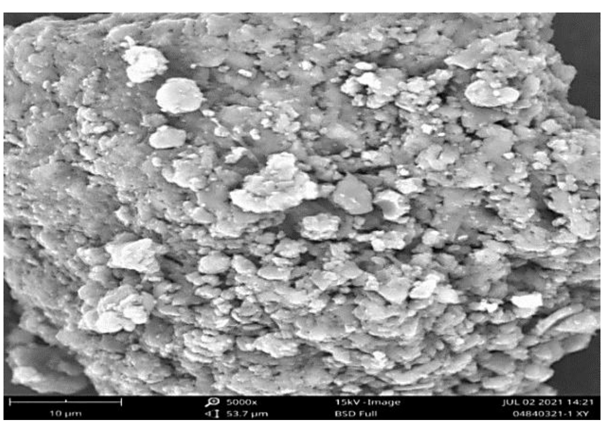
In the final coal, there was the emergence of the C-H group in the absorption area of 1,446.38 $cm^{-1}$. The presence of O-H and C-H groups were supported by the presence of carbonyl groups (C-O). It is known that carbonyl groups had an absorption area at wavelengths of 1,650-550 $cm^{-1}$ (Anam, Sirojudin and Firdausi, 2007). In the initial coal sample, the C-O group had an absorption band in an area of 1,175.37 $cm^{-1}$ with an intensity of 61,992 %T. In the final sample, the C-O group was seen around 1,266.59 $cm^{-1}$ with an intensity of 39,660 %T and in the final sample also appeared compounds of the C-O carbonyl group of 536.33 $cm^{-1}$ with an intensity of 47.207 %T.

The content of alkene compounds in coal were seen in the presence of the C=C function group in the FTIR test. It was seen in the wavelength number of 1,614.77 $cm^{-1}$ in the initial sample and 1,613.91 $cm^{-1}$ in the final sample. The C=C functional group decreased in intensity at 58.804 %T in the initial sample then decreased in the final sample of 35,928 %T. This is according to the statement of Skoog et al. that the group C=C alkene was around 1680-1610 $cm^{-1}$ (Skoog, Holler and Crouch, 2017).

## SEM-EDX Analysis

The use of heavy oil as additives could be adsorbed to coal surfaces and pores. With SEM analysis we could found out the surface conditions and pores of coal that show the topographic results of samples with bright colours that represent higher surfaces and dark colours representing lower surfaces (Martinez, 2012). The composition of the element was known through the EDX test. EDX analysis was used to analyze the quantitative percentage of each element (Lowinsky *et al.*, 2021). The results analyzed for SEM-EDX were dewatered coal samples with a ratio of 100: 45 (g/mL) at a dewatering slurry temperature of 140 $^oC$. Based on Figure 9, the number of dark areas decreases after the dewatering slurry process. It can be stated that the heavy oil covers the pores of coal.

Based on the results of the EDX analysis as shown in Table 2 and Figure 10, there was a decrease and increase in the concentration of elemental weight from the initial sample of coal and the final sample of coal. However, only a small portion of the elements showed



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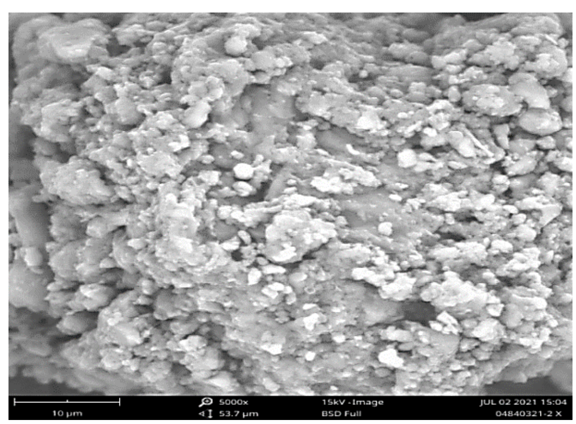


b.

Figure 9. Coal Morphology (a) before treatment and (b) after treatment

a decrease in weight concentration in the final sample. The elements that showed a decrease in weight concentration only occurred in the element O, while other elements such as C, Ca, Mg, Al, and Si showed an increase in weight concentration. It was comparable to the increase in calorific value that occurs in coal after upgrading because the carbon in coal has increased (Muchjidin, 2006).

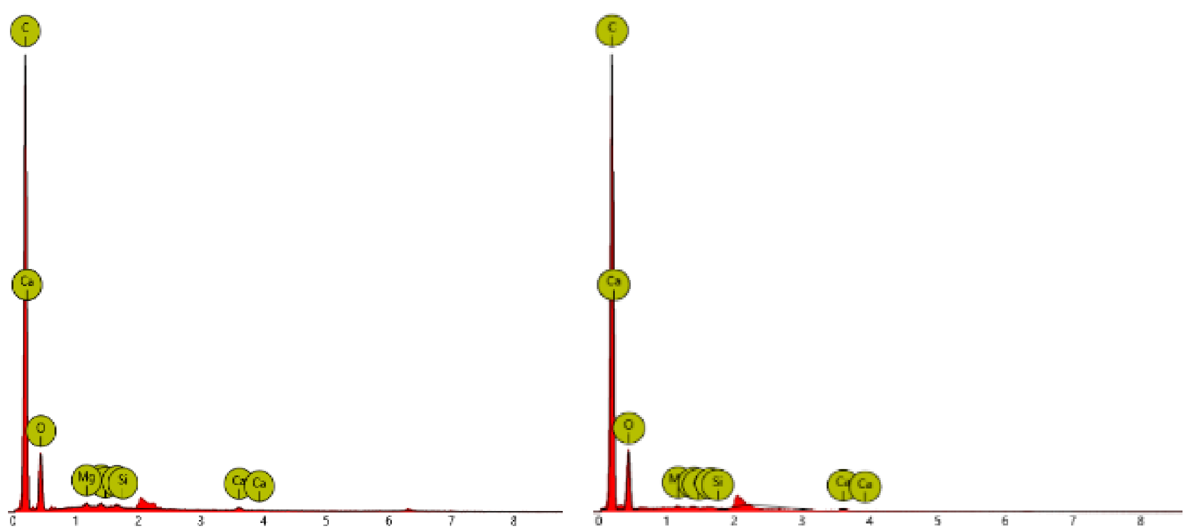
Table 2. Concentration of Elemental in Coal

| Element | Coal before treated | | Coal after treated | |
|---|---|---|---|---|
| | atom (%) | weight (%) | atom (%) | weight (%) |
| C | 78.80 | 73.37 | 79.42 | 73.93 |
| O | 20.88 | 25.90 | 20.11 | 24.94 |
| Ca | 0.09 | 0.28 | 0.18 | 0.56 |
| Mg | 0.11 | 0.20 | 0.11 | 0.20 |
| Al | 0.07 | 0.14 | 0.11 | 0.22 |
| Si | 0.05 | 0.11 | 0.07 | 0.15 |

Based on the results of the EDX analysis as shown in Table 2 and Figure 10, there was a decrease and increase in the concentration of elemental weight from the initial sample of coal and the final sample of coal. However, only a small portion of the elements showed a decrease in weight concentration in the final sample. The elements that showed a decrease in weight concentration only occurred in the element O, while other elements such as C, Ca, Mg, Al, and Si showed an increase in weight concentration. It was comparable to the increase in calorific value that occurs in coal after upgrading because the carbon in coal has increased (Muchjidin, 2006).

## CONCLUSION

The addition effect of heavy oil from catalytic cracking of Polypropylene-Low Density Polyethylene plastic waste pyrolysis oil on improving the quality of lignite has been studied. The heavy oil added to coal with a ratio of 45:100 with a temperature of 140 ºC were the optimal process condition in slurry dewatering that increased the calorific value of coal from 3,662 to 6,374 cal/g or increase to 74.06 %. The heavy oil added to coal with a ratio of 30:100 with a temperature of 150 ºC were the optimal process condition in slurry dewatering can increase the calorific value of coal from 3,662 to 6,168 cal/g or increase to 68.43 %. The use of heavy oil as an additive could filled the pores of coal so that the moisture content in coal was reduced and the calorific value of coal was increased.




a. b.

Figure 10. EDX spectra of (1) coal before treated and (b) coal after treated

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