Low-rank coal and poly fatty acid distillate characterization as a preparation of coal upgrading palm oil technology

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Abstract. The utilization of low-rank coal is restricted by such factors as high moisture content, low heating value, high propensity to low-temperature oxidation, spontaneous combustion, etc. Some coal upgrading technologies to reduce the moisture content have been developed, one of them is coal upgrading palm oil technology using palm fatty acid distillate as an additive to keep the stability of moisture content in the coal after the process. To study the possibility of the upgrading technology application in Indonesia, some studies have been conducted. The study covered coal characterization such as proximate, ultimate and calorific value, palm fatty acid distillate for stabilization of upgraded low-rank coal and coal upgrading by coal upgrading palm oil technology in laboratory scale. By using 7 Indonesian low-rank coals and 4 palm fatty acid distillates, it is confirmed that the coal upgrading palm oil technology is effective to reduce the moisture content and increase the calorific value of low rank coal.

1. Introduction

Indonesia continues to be a significant player in the global coal mining industry although profit margins of miners are dwindling due to the swelling production costs coupled with the low commodity prices amid continuing global economic uncertainties. Indonesia remains the world’s largest exporter of thermal coal, shipping about 80% of the country’s production. In 2019, the Country produced 616 million tons of coal, in which the use domestically of 22.44% and export of 77.56%. According to the Geology Agency, 2018 Indonesia's coal resources are 151.4 billion tons and total reserves of 39.89 billion tons [1], ranking the sixth largest in the world.

In 2014, the House of Representatives passed a regulation on National Energy Policy proposed by the National Energy Board (DEN). The regulation, which governs the country’s energy strategy and policy for the 2013-2050 period, stipulates the targets of primary energy diversification, which will reduce the role of oil in the energy mix from about 50% at present to about 24% by 2025 and to less than 20% by 2050 [2]. Similarly, coal’s portion will also, however, be cut from 30% in 2025 to 25% in 2050. Meanwhile, the role of natural gas will be increased from 20% at present to 22% by 2025 and 24% by 2050 and renewable energy from 6% to 23% by 2025 and 31% by 2050.

The utilization of LRC is restricted by such factors as high moisture contents, low heating value, high propensity to low-temperature oxidation, spontaneous combustion, etc. [3]. Upgrading low-rank coal (LRC) in order to reduce the moisture content is an interesting work in the coal research area, since
this effort can suppress the low-temperature oxidation, self-heating, and spontaneous combustion during transportation and storage [4].

Coal upgrading continues to be a major priority within the industry with a significant emphasis being placed on the need to obtain maximum performance from current unit operations, in particular to reduce product moisture to levels competitive with those of overseas producers. Coal upgrading also improves understanding of the major parameters that affect its process, so that maximum machine performance can be obtained and to identify cost-effective techniques that can be conveniently retrofitted into existing plants to improve dewatering and reduce product moisture [5]. A further major driver, and one not restricted to upgrading, is the need to minimize circuit complexity and the costs associated with maintenance [4].

There are two purposes for LRC upgrading. One is to use LRC directly for power plants. LRC is dried by using waste heat such as flue gas and the dried coal is used for boilers. This increases the efficiency of the boiler and reduces the amount of waste gas and ashes, which leads to a decrease in the facility load in the power plants. The other is to convert the LRC into a transportable state at a LRC mine for export. LRC has a high spontaneous combustion property and it is hard to store or transport, unless it is used in a boiler immediately after being dried. Therefore, it is necessary to change the raw LRC into a stable and transportable state [6].

Some of the coal upgrading technologies have been discussed elsewhere. Among them were steam/hot water drying [4,7–9], upgraded brown coal [10–12], circulating fluidized bed drying [13–15], microwave drying [16–18] Combdry drying [6,19–21], rotary tube drying [18,22] etc. with their advantages and some obstacles to be applied in commercial scale. Therefore, until now the application of such upgrading technology in Indonesia is still very limited. The limitation of coal upgrading application in commercial scale is affected by various conditions such as technology, market, economics, environment, policy and strategic issues being prime. At the present time, coal upgrading primarily entails drying prior to direct use as fuel for combustion boilers in power plants to reduce the surface moisture of the coal.

One of the coal upgrading technologies is coal upgrading palm oil (CUPO) that is developed by the Korean Institute of Energy Research (KIER), Korea. It uses a steam tube dryer (STD) as a drying device and palm residues such as poly fatty acid distillate (PFAD) and palm acid oil (PAO) as coating materials [19]. The palm residues are solid at room temperature, liquid around 60°C and have low viscosity. They have a high affinity with coal, so they are easily mixed with coal during drying and become solid after drying, serving as a binder. It is confirmed that The CUPO process upgrades a LRC and reduces moisture content to be equal with a high rank coal [23]. Therefore, it is expected to increase the combustion efficiency and decrease the susceptibility to spontaneous combustion. Schematic process of CUPO is shown in Figure 1 [19].

![Figure 1. Schematic process of CUPO.](image)
PFAD is produced as a byproduct of palm and palm kernel oil. Indonesia is one of the largest palm oil producing countries, supplying almost half of the world’s palm oil. Indonesia produces about 3 million tons of crude palm oil (CPO), becoming the largest CPO producer in the world with a total production of 32 million tons or about 46.6% of the world’s total CPO production. World market demand for CPO increases continuously, in 2020 is estimated to reach 95.7 million tons. Meanwhile, the development of the Indonesian palm oil industry shows a positive trend with an increase in production every year. Therefore, it is important to recognize the potential of palm oil waste as an added value to the Indonesian palm oil industry [24, 25].

Palm oil or palm kernel oil produces edible and non-edible products. The edible products produce cooking oil/fats, fats for bakery, margarine and specialty fats. While the non-edible products produce oleochemical and soap. With an advanced process, the oleochemical produces fatty acid, fatty alcohol and glycerin. The downstream product of palm and palm kernel oil is presented in Figure 2 [26].

![Figure 2. The downstream of palm oil and palm kernel oil products.](image)

The objective of this study was to study the characteristics of some LRCs and residual palm oil of PFAD in such areas that are close to the LRC deposits. As a preliminary study, the characteristics of LRC after upgrading by CUPO technology in laboratory scale were limited to moisture and calorific value analyses. The aim of this characterization was to identify the degree of dewatering and the calorific value improvements due to upgrading by CUPO technology. More detailed research to study the influencing factors due to the upgrading process such as volatile matter, fixed carbon including combustion properties of the upgraded coal is still in progress.

2. Materials and Method

2.1. Characterization of LRC
The study was carried out by using 7 LRCs come from Meulaboh (Aceh), Sekayan (North Kalimantan), Tabang (East Kalimantan), Pendopo (South Sumatera), Banko (South Sumatera), Mulia (South Kalimantan) and Sorong (West Papua). LRC characterization is carried out through proximate analysis of inherent moisture, ash, volatile matter and fixed carbon, ultimate analysis of carbon, hydrogen, nitrogen, total sulfur and oxygen and calorific value analysis. Based on the calorific value in air dried basis (adb), the quality of coal is divided into 4 groups as can be seen in Table 1 [27].
Table 1. Coal quality based on caloric value cal/g in air dried basis.

| Calorific value in air dried basis, kcal/kg | Category               |
|-------------------------------------------|------------------------|
| < 5,100                                   | Low quality coal       |
| 5,100-6,100                               | Medium quality coal    |
| 6,100-7,100                               | High quality coal      |
| >7,100                                    | Very high quality coal |

2.2. Characterization of PFAD

There are 4 PFAD samples used in this study that come from Palm Oil Research Centre (PORC) Medan, North Sumatera, Sinar Mas, South Sumatera and South Kalimantan and Wilmar Surabaya, East Java. The characterization includes caloric value, melting point, percent of impurities, free fatty acid (FFA) as palmic, moisture content, and fatty acid composition.

2.3. Upgrading of LRC

LRC upgrading by CUPO technology was conducted by using 7 LRCs and 4 PFADs samples as mentioned above. The pulverized coal sample (less than 75 µm) was mixed with PFAD in different concentrations of 1, 5 and 10% according to the PFAD variables and the resulting coal and PFAD mixture was placed in a 50 g petry disk and was put in an oven. Nitrogen at a flow rate of 80 cm³/min was passed through a flow meter into the oven. The oven was heated at a rate of 3-4°C/min to a desired heat treatment temperature (110-120 °C) and the temperature was kept for 12 hours [7]. The petry disk was pulled out and cooled. Moisture content was determined by establishing the loss in weight of the sample after heating [28]. The degree of dewatering and calorific value improvement is defined as in equation 1 and 2 respectively.

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\text{Degree of dewatering:} \quad \frac{\text{(M)} \text{ raw coal} - \text{(M)} \text{ upgraded coal}}{\text{(M)} \text{ raw coal}} \times 100\% \quad (1)
\]

\[
\text{The calorific value improvement:} \quad \frac{\text{(CV)} \text{ upgraded coal} - \text{(CV)} \text{ raw coal}}{\text{(CV)} \text{ raw coal}} \times 100\% \quad (2)
\]

Where M is the moisture content (%) and CV is calorific value (cal/g) of the coal on an air dried basis.

3. Results and Discussions

3.1. LRC Characterization

The proximate, ultimate and caloric value analyses of some LRC samples have been conducted to know the characteristics of the coals. The results are presented in Table 2. The total moisture of Pendopo coal, South Sumatera is the highest compared to other LRCs i.e., 61.25% and the inherent moisture of 20.91%. Consequently the caloric value is also the smallest i.e., 4,603 cal/g (ADB). This coal faces some problems if it is used as fuel, among others are high cost during handling and transportation [29], spontaneous combustion tendency and low combustion efficiency so that a lot of coal should be burned to obtain a certain energy, resulting in high CO emissions [30]. With a relatively low sulfur content (0.18%), this coal still can be utilized as fuel if it is upgraded prior to being used.
Table 2. Proximate, ultimate and calorific value analyses results.

| Analysis                      | Meulaboh | Sekayan | Tabang | Pendopo | Banko | Mulia | Sorong |
|-------------------------------|----------|---------|--------|---------|-------|-------|--------|
| Total moisture wt% (AR)       | 36.81    | 45.42   | 35.78  | 61.25   | 30.13 | 36.00 | 42.04  |
| Calorific value, cal/g (ADB)  | 5,009    | 5,379   | 5,194  | 4,603   | 5,839 | 5,191 | 4,823  |
| Proximate (ADB), wt%          |          |         |        |         |       |       |        |
| Inherent Moisture             | 15.91    | 12.55   | 20.36  | 20.91   | 17.36 | 17.31 | 20.55  |
| Ash                           | 5.32     | 5.41    | 3.03   | 5.31    | 1.91  | 4.30  | 4.22   |
| Volatile matter               | 42.01    | 45.18   | 37.60  | 38.06   | 40.41 | 41.42 | 38.66  |
| Fixed carbon                  | 36.76    | 36.86   | 39.01  | 35.72   | 41.32 | 36.97 | 36.57  |
| Ultimate analysis (DB), wt%   |          |         |        |         |       |       |        |
| Carbon                        | 53.64    | 57.1    | 54.66  | 51.18   | 62.64 | 53.87 | 51.23  |
| Hydrogen                      | 5.51     | 5.44    | 5.92   | 5.97    | 6.31  | 5.95  | 5.99   |
| Nitrogen                      | 0.89     | 0.76    | 0.74   | 0.54    | 0.89  | 0.73  | 1.15   |
| Sulfur                        | 0.12     | 0.34    | 0.09   | 0.16    | 0.18  | 0.13  | 0.34   |
| Oxygen                        | 34.52    | 30.95   | 35.56  | 36.84   | 29.07 | 35.02 | 37.07  |

Note: AR: As Received; ADB: Air Dried Basis; DB: Dry Basis

According to the coal grouping based on calorific value (Table 1), the Sekayan, Tabang, Banko and Mulia coals are categorized as medium quality coals, while the Meulaboh, Pendopo and Sorong coals are categorized as low quality coals. All samples show very low ash content and it is very beneficial as fuel for power generation since it generates very low ash that leads to easiness/less complicated handling. The volatile matter, fixed carbon and ultimate analyses results as well, are shown as a general characteristic of LRC which are relatively high in volatile matter, low fixed carbon and high oxygen contents [31].

3.2. PFAD characterization

Oil palm plantations and palm oil processing mills are no longer confined to the island of Sumatra but have expanded to Kalimantan (Borneo), Sulawesi (Celebes) and Papua. Currently, almost 70 percent are located on Sumatra [32]. The remainder around 30 % is largely found in Kalimantan. The result of PFAD characterization can be seen in Table 3.

Table 3. PFAD characteristics.

| Parameter                        | PORC, North Sumatra | Sinar Mas, South Sumatra | Sinar Mas, South Kalimantan | Wilmar, East Java |
|----------------------------------|---------------------|--------------------------|-----------------------------|------------------|
| Gross calorific value, cal/g     | 9,328               | 9,317                    | 9,369                       | 9,403            |
| Melting point, °C                | 46.00               | 47.00                    | 45.00                       | 45.00            |
| Impurities,%                     | 0.01                | 0.01                     | 0.01                        | 0.01             |
| FFA as palmitic, %               | 81.06               | 88.42                    | 91.24                       | 88.95            |
| Moisture content, %              | 0.34                | 0.08                     | 0.32                        | 0.20             |
| Fatty acid composition           |                     |                          |                             |                  |
| Lauric acid C12:0, %             | 4.10                | 2.91                     | 2.84                        | 1.80             |
| Myristic acid C14:0, %           | 5.92                | 5.09                     | 3.15                        | 3.80             |
| Palmitic acid C16:0, %           | 62.83               | 66.17                    | 61.95                       | 62.29            |
| Stearic acid C18:0, %            | 2.04                | 1.89                     | 2.54                        | 2.54             |
| Oleic acid C18:1, %              | 18.98               | 18.39                    | 22.04                       | 22.65            |
| Linoleic C18:2, %                | 4.34                | 4.53                     | 5.93                        | 5.99             |

The characteristics of the PFAD that have been collected from some areas in Indonesia show a similarity between one to each other. High calorific value (>9,300 cal/g), low melting point. Low
impurities and low moisture content indicate that the PFAD can be used as an additive to coat the coal pore after the heating process so that the moisture content of the coal will be stable.

3.3. LRC upgrading
The upgrading of LRC and PFAD mixtures have been conducted by using 7 LRCs and 4 PFADs samples. The mixing of LRC and PFAD samples are carried out in such a way based on the proximity of the coal and PFAD locations. The degree of dewatering and calorific value improvement can be seen in Table 4. The effect of PFAD addition on inherent moisture content and degree of dewatering are shown in Figure 3 and 4 respectively. While the effect of PFAD addition on calorific value and calorific value improvement are shown in Figure 5 and 6.

Table 4. LRC and PFAD mixture.

| LRC Origin      | PFAD Origin       | PFAD in LRC wt% | Moisture content, wt % | Degree of dewatering % | Calorific value cal/g | CV Improvement % |
|-----------------|-------------------|-----------------|------------------------|------------------------|-----------------------|------------------|
| Meulaboh, Aceh  | Medan North Sumatra| 1              | 12.01                  | 24.51                  | 5,397                 | 7.75             |
|                 |                   | 5              | 11.98                  | 24.70                  | 5,545                 | 10.70            |
|                 |                   | 10             | 11.21                  | 29.54                  | 5,772                 | 15.23            |
| Sekayan, North Kalimantan | South Kalimantan | 1              | 10.29                  | 18.01                  | 5,655                 | 5.13             |
|                 |                   | 5              | 9.96                   | 20.64                  | 5,783                 | 7.51             |
|                 |                   | 10             | 9.61                   | 23.43                  | 6,005                 | 11.64            |
| Tabang, South Kalimantan | South Kalimantan | 1              | 16.04                  | 21.22                  | 5,779                 | 11.26            |
|                 |                   | 5              | 15.50                  | 23.87                  | 5,991                 | 15.34            |
|                 |                   | 10             | 15.23                  | 25.20                  | 6,267                 | 20.66            |
| Pendopo, South Sumatera | South Sumatra | 1              | 18.08                  | 13.53                  | 5,163                 | 12.17            |
|                 |                   | 5              | 17.29                  | 17.31                  | 5,393                 | 17.16            |
|                 |                   | 10             | 16.86                  | 19.37                  | 5,730                 | 24.48            |
| Banko, South Sumatera | South Sumatra | 1              | 11.88                  | 32.50                  | 6,446                 | 10.40            |
|                 |                   | 5              | 11.64                  | 33.86                  | 6,645                 | 13.80            |
|                 |                   | 10             | 10.81                  | 38.58                  | 6,912                 | 18.38            |
| Mulia, South Kalimantan | South Kalimantan | 1              | 15.61                  | 9.82                   | 5,808                 | 11.89            |
|                 |                   | 5              | 14.40                  | 18.20                  | 6,022                 | 16.01            |
|                 |                   | 10             | 14.16                  | 18.89                  | 6,219                 | 19.80            |
| Sorong, West Papua | Surabaya East Java | 1              | 17.36                  | 15.52                  | 5,570                 | 15.49            |
|                 |                   | 5              | 16.72                  | 18.64                  | 5,704                 | 18.27            |
|                 |                   | 10             | 16.48                  | 19.81                  | 6,208                 | 28.72            |
From Figure 3 it can be seen that the increase of PFAD content decreases the moisture content, so that degree of dewatering increases with the increase of PFAD content in the coal (Figure 4). The highest degree of dewatering is reached by Banko coal which was mixed with PFAD and comes from South Sumatera of 10%. The degree of dewatering reached 38.58%. At this condition (PFAD 10%), the lowest degree of dewatering is reached by Mulia coal, South Kalimantan which was mixed with PFAD from South Kalimantan i.e. 18.89% (Table 2).

According to Yu, et al, 2014 [30], classification of water in coal involves (1) Interior adsorption water is contained in micropores and microcapillaries within each coal particle, deposited during coal formation; (2) Surface adsorption water forms a layer of water molecules adjacent to cold molecules but on the particle surface only; (3) Capillary water is contained in capillaries; (4) Interparticle water is contained in small crevices found between two or more particles; and (5) Adhesion water forms a layer or film around the surface of individual or agglomerated particles. Water types (4) and (5) (i.e. surface moisture) can be removed by mechanical dewatering methods. Water type (3) can be removed partially, depending upon the size of the openings in the coal surface and the drying temperature. Water types (1) and (2) are inherent moisture and can be removed by thermal drying processes.
Based on the above statement, the significant increase of degree of dewatering of Banko coal compared to Mulia coal might be due to the moisture in Banko coal is categorized as water type (3), while the water in Mulia coal is categorized as water type (1) or (2) that should be removed by thermal drying at a higher temperature than the upgrading temperature of CUPO technology which is only 120°C. Thus, in order to evaporate water that is inside a large stack of coal, the coal is heated to a higher temperature and heat is conducted to the water trapped inside [33]. In addition, LRCs contain many microporous structures. Water stored in these pores of different sizes forms different structured clusters, water adsorbed in some micropores is difficult to remove using conventional methods [3].

The moisture content from coal is removed by three ways: the thermal influences, the CO₂ produced due to the thermal damage of functional groups, and thirdly because of the collapse and shrinkage influences of coal [34]. The CUPO technology is carried out at a relatively low temperature of 110-120°C. The decrease of inherent moisture most likely only due to the thermal influence.

![Figure 5. Effect of PFAD addition on calorific value.](image1)

![Figure 6. Effect on PFAD addition on calorific value improvement.](image2)
Figure 5 shows the calorific value of upgraded coal samples. The calorific value increases with the increase of PFAD, as well as the calorific value improvement (Figure 6). The highest calorific value improvement is reached by Sorong coal i.e. 28.72% at the addition of PFAD of 10%. It could be understood because the Sorong coal was mixed with PFAD that has high calorific value, which is the highest compared to the other PFAD samples. The increase in calorific value improvement with the addition of 5% to 10% PFAD is very significant.

Banko coal which has the highest degree of dewatering compared to other coal samples, the calorific value improvement is not as high as Sorong and Pendopo coals which were relatively low degrees of dewatering. However, the calorific value of Banko coal remains the highest compared to other upgraded coals. It shows that the calorific value improvement does not only depend on degree of dewatering [35] but also on other matters such as elemental analysis (carbon, hydrogen and oxygen), ash contents [36]. Karthikeyan et al. [33] reported that for LRCs (whose volatile matter content is above 35%), the calorific value will increase with a decrease in volatile matter content. While in this preliminary study the elemental compositions, ash and volatile matter after the upgrading process were not analyzed. Hence in further research, it is important to analyze the change in coal properties after drying.

Another evaporation upgrading technology of dual stage drying which was also used Pendopo coal, South Sumatera, the optimum degree of dewatering was 77.27% at the coal particle size of < 9.5 mm [22] is much higher than that of this CUPO upgrading. It could be understood because the average temperature of the process was approximately 130-180°C, while the CUPO technology was held at temperature of 110-120°C, higher temperature led to higher degree of dewatering [6,37]. At least two classes of water exist in LRC at any particular temperature. Firstly, water which can be removed by evacuation at temperature of < 150 °C and secondly chemisorbed water which can be released only by raising the temperature (>150 °C) to cause thermal decomposition of functional groups [30].

4. Conclusion

Indonesian coal mostly is classified as LRC with high moisture content and low calorific value. By coal upgrading using CUPO technology that is developed by KIER, the quality of coal can be enhanced. The palm oil industry is an important part of the national economies of Indonesia. Likewise, Indonesia is also the world’s largest producer of crude palm oil in the world. By using 7 Indonesian LRCs and 4 PFADs, the increasing of PFAD content increases the degree of dewatering and the calorific value improvement. It is confirmed that CUPO technology is effective to reduce moisture content and increase the calorific value of LRC. With in-depth research at a higher scale, it is hoped that CUPO technology can be immediately applied on a commercial scale so that the utilization of low rank coal is optimal.

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