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STUDY ON UPGRADED LOW RANK COALS PROPERTIES

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ABSTRACT

Upgraded brown coal (UBC) process has been discussed elsewhere. This process has been developed to produce an upgraded low rank coal with quality similar to a bituminous coal which is acceptable commercially and has low moisture content. Three Indonesian low rank coals, Berau, Tabang and Samaranggau coals were upgraded by upgraded brown coal (UBC) process to study the influence of the process on the properties of the upgraded low rank coals by conducting chemical and physical analyses such as proximate, ultimate including calorific value and equilibrium moisture, functional group of C-H and C=O, coal petrography, specific surface area and briquettability.

The result of proximate analysis indicated that the inherent moisture of the upgraded low rank coals decreased significantly compared with that of the raw coals. Hence, the calorific value of the upgraded coals increased. The ash content of the upgraded coals was not change obviously due to that the UBC process was conducted at low temperature. However, the volatile matter content increased slightly due to the kerosene or residue that left and pluged over coal pores to prevent the reabsorbtion of moisture. From ultimate analyses, carbon content of the upgraded coals increased, whereas the hydrogen and oxygen contents decreased. The UBC process hardly affected the sulfur and nitrogen contents. The result of equilibrium moisture measurement showed that the moisture content of all upgraded coals were less than 9%. The functional groups of C-H and C=O of the upgraded coals were slightly less than those of the raw coals. The aromaticity of the upgraded coals were increased. The petrography of both the raw and the upgraded coals indicated that the mean vitrinite reflectance was slightly higher in the upgraded coal compared to the raw coal. There was no significant quantity and textural differences of maceral in both coals. The specific surface area of the upgraded coals was lower than that of the raw coal due to the plugging of pore structure and shrinkage by residual oil addition. The upgraded low rank coals briquette according to drop shutter and compressive strength tests indicated good characteristics of briquette.

Based on these results, UBC process only reduces the moisture content, so that the calorific value of the coal increases. Whereas the other parameters are not significantly change. UBC process does not increase the rank of the coal, therefore, it could only be applied to improve the calorific value of low rank coal which has low ash and sulfur contents.

Keywords: low rank coal, UBC, upgraded coal properties

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INTRODUCTION

Coal deposits are widely distributed throughout the Indonesian archipelago with total resources amounting to 93.4 billion tons, which are classified as 58% of lignite, 27% of subbituminous, 14% of bituminous and small amount (less than 1%) of anthracite. Lignite and subbituminous coal have high moisture and volatile matter contents, low calorific value and high oxygen content, so it is uneconomic to carry over long distance transportation. In addition, precaution has to be done to prevent spontaneous combustion from the coal which is dried partially. One of key questions is whether a low rank coal (LRC) can be upgraded in the form of energy, which can be transported economically and storaged safely.

Several methods of dewatering and upgrading processes to reduce moisture content and producing coal with higher calorific value and lower transportation cost have been studied since 1920s (Suwono et al., 1999). Among of them, the UBC (upgraded brown coal) process which has been developed by Kobe Steel Ltd., Japan will be applicable (Shigehisa et al., 2000). The UBC process upgrades a LRC and reduces moisture content to be equal with a high rank coal. Therefore it is expected to decrease the susceptibility to spontaneous combustion and easy to be transported elsewhere. This paper reports the properties of upgraded low rank coals by UBC process compared with the raw low rank coals. Three samples coming from East Kalimantan, have been processed to study the change that caused by UBC process. In particular, this paper reports on chemical and physical properties such as the proximate and ultimate analyses, the calorific value, the equilibrium moisture, coal petrography, functional group of C-H and C=O, specific surface area and coal briquettability test such as drop shutter and compressive strength tests. Combustion characteristics of the coals were presented in previous paper (Umar et al., 2005)

The objective of this study is to provide the general knowledge which related to the UBC process as a basic information on both the raw and the upgraded coals characteristics in order to establish the advantages of the process from the technical points of view. This study is very valuable in helping to make decisions in selecting upgrading process to increase the low rank coal utilization.

EXPERIMENTAL

The raw coals coming from 3 coal mining areas in Indonesia, Berau, Tabang and Samaranggau were processed in UBC pilot plant in Palimanan, Cirebon. The UBC process consists of five main sections; they are coal preparation, slurry dewatering, coal-oil separation, oil recovery and coal briquetting (Daulay et al., 2003). The UBC process has been described elsewhere (Deguchi et al., 2002). The raw and the upgraded coals were analyzed to study the change in chemical and physical characteristics due to the process. The proximate and ultimate analyses including equilibrium moisture and calorific value in air dried basis (adb) were conducted according to the ASTM Standard 2005.

A functional group analysis was performed using a Fourier transform infra red HORIBA FT-720 spectrometer (FTIR) with the resolution of 1 cm^{-1} to investigate the change of quality due to the upgrading by UBC process. Sampling was carried out by the KBr pellet method and measurements conducted in selected spectra range of 3000-2700 cm^{-1} and 1800-1500 cm^{-1}. Those ranges were assigned to the aliphatic hydrogen (C-H functional group) and oxygen containing structures (C=O functional group), respectively (Ohki et al., 1999). Coal petrography analysis was carried out by polarization microscope MPM-200. Petrography characteristics of coal can be expressed in terms of two essentially independent concepts, i.e. coal type and coal rank. Coal type refers to the nature of the organic matter found in the coal and coal rank refers to the stage of coalification that was reached by the organic matter. Therefore, petrographic study of coal is based on the morphology, colour, size of the constituents, reflectance and anisotropy of the macerals in reflected white light and autofluorescence of the constituents under blue-ultraviolet radiation. This research intends to investigate if the upgraded coal can be distinguished petrographically from the raw coal.

To study the physical properties of the upgraded low rank coals, the specific surface area was measured by BET (Brunauer Emmet Teller) method using CO_2 at 25°C, by using SHIBATA APP.SA-100 based on the amount of nitrogen absorbed by the solid particles surfaces. The nitrogen flow rate was 150 ml/min (Mahidin et al., 2003). Before subjected to the test, coal samples were dried in nitrogen atmosphere at 180°C. The specific surface area of the coal in cm^2/g could be obtained. To study the briquettability of the upgraded low rank coals drop shutter and compressive strength tests were conducted.

RESULT AND DISCUSSION

Proximate and ultimate analysis

The results of proximate and ultimate analysis including equilibrium moisture and calorific value of the raw and the upgraded coals are shown in Table 1.

The main goal of the upgrading process is to reduce moisture content of the low rank coal, so that the calorific value is increased. The parameter of dewatering is evaluated by the moisture content data of the raw and upgraded coals. The degree of dewatering is defined by the formula of:

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

The inherent moisture content of Berau upgraded coal decreased significantly from 16.13 to 1.52% or degree of dewatering was 90.58%, whereas the value for Tabang and Samaranggau upgraded coals were 66.19% and 79.18% respectively.

The calorific value of the Berau coal increased from 5,324 up to 6,805 kcal/kg or increased 27.82%, whilst Tabang and Samaranggau coals increased 21.98% and 25.00% respectively. It can be seen that the decreasing of inherent moisture is linier to the increasing of calorific value. Berau coal has the highest degree of dewatering resulting the highest increase of the calorific value, followed by Samaranggau and Tabang coal. Figures 1 and 2 illustrate the decrease of moisture contents and increase in calorific value of the upgraded coals compared with those of the raw coals, respectively.

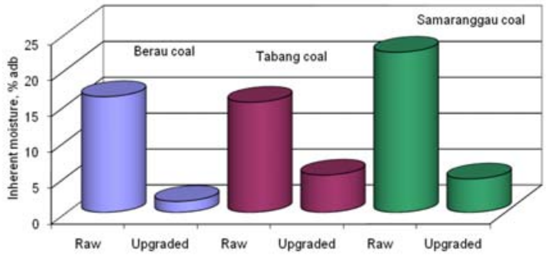

Figure 1. Moisture content of the raw and the upgraded coals

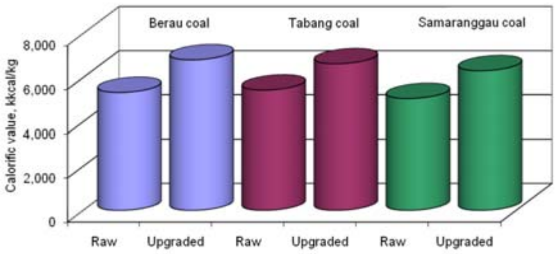

Figure 2. Calorific value of the raw and the upgraded coals

Table 1. Proximate and Ultimate analysis results of the raw and upgraded coals

Coals	Berau		Tabang		Samaranggau	
Analysis, air dried basis	Raw	Upgraded	Raw	Upgraded	Raw	Upgraded
Inherent Moisture, %	16.13	1.52	15.35	5.19	22.33	4.65
Ash, %	6.36	6.81	4.42	3.93	2.15	2.61
Volatile matter, %	37.20	46.78	40.68	48.34	38.15	47.67
Fixed carbon (diff), %	40.31	44.89	39.55	42.54	37.37	45.07
Equilibrium Moisture, %	-	5.08	-	6.60	-	8.48
Sulfur, %	0.56	0.52	0.14	0.18	0.10	0.16
Carbon, %	57.01	65.19	57.55	66.58	53.60	64.43
Hydrogen, %	6.57	4.80	6.06	5.07	6.19	5.30
Nitrogen, %	1.60	1.15	0.77	0.88	0.69	0.86
Oxygen (diff), %	27.90	21.53	31.06	23.36	37.27	26.64
Calorific value , kcal/kg	5,324	6,805	5,431	6,625	5,048	6,310

where: M is the moisture content (%) of the coals in air dried basis

Based on the equilibrium moisture (EM) content, all of the upgraded low rank coals had EM content less than 9%. The ash contents of the upgraded coal were not obviously changed. Berau and Samaranggau upgraded coals were slightly higher compared to that of the raw coals, except that of Tabang coal which slightly decreased. The increase of ash content of the upgraded low rank coals related to the decreasing of moisture content due to the UBC process was caused by the increasing of the total coal mass. In the other hand, the decrease of ash content of the upgraded Tabang coal, was probably related to the dissolution of impurities in the coal by kerosene during coal treatment. The impurities type in Tabang coal was different from the other coals. It can be explained that the impurities in Tabang coal were easy to be removed because most of the impurities were extraneous type (Umar et al, 2005). However, in general, the change in ash content in the upgraded coals compared to the raw coals was not significant. On the other hand, the volatile matter of all of the upgraded low rank coals increased. The increase of volatile matter was also caused by kerosene or residue that left and coated the surface of the upgraded coal in which the kerosene or residue was detected as volatile matter when the coal was burned. The increase of fixed carbon was certainly related to the reduction of the moisture content.

Based on the ultimate analysis, the carbon content of the upgraded low rank coals significantly increased with increasing of fixed carbon. The increase of carbon content will improve the combustibility of the coal. It means, the utilization of the coal as a fuel will be enhanced. The removing of water from coal causes the decrease of hydrogen and oxygen contents in the upgraded coals. While total sulfur and nitrogen contents were slightly changed. The increase of sulfur content in some of the upgraded coals was mostly related to the increasing of the coal mass due to the decreasing of moisture content.

Functional Group Analysis

The FTIR spectrum for the raw and upgraded coals is shown in Figure 3. The selected ranges of 3000-2750 cm^{-1} and 1800-1500 cm^{-1} correspond to the structure of aliphatic hydrogen (C-H functional group) and the oxygen containing structures (C=O functional group) respectively. The zone of aliphatic hydrogen corresponded to asymmetric methyl ($-CH_3$) and methylene ($-CH_2-$), methane (C-H) and symmetric methyl ($-CH_3$) and methylene ($-CH_2-$), were observed in 2965, 2920, 2895, 2870 and 2850 cm^{-1}, respectively. The zone of 1800-1500 cm^{-1} corresponds to carboxyl group and quinines (esters, aliphatic and aromatic COOH, conjugated and highly conjugated C=O), aromatic carbon (aromatic C=C), and carboxylic groups (COO-) stretching at 1770, 1700, 1655,1615, 1580, 1560 and 1540 cm^{-1}, respectively (Ibara and Miranda, 1996).

Comparing the peaks of C-H and C=O group of the raw and the upgraded low rank coals, the peaks of the upgraded low rank coals was slightly lower than those of the raw coals. It is according to the fact that the hydrogen and oxygen contents of the upgraded coals are almost equal or slightly less than those of the raw coals as shown in Table 1. The slightly decrease of hydrogen and oxygen contents caused by the UBC process was conducted under mild condition (relatively low temperature and low pressure).

The parameters of the raw and upgraded low rank coals are summarized in Table 2. The following parameters defined as the ratio of deconvoluted peak area, RCH_3/CH_2, R*ar/al*, R*COOH/ar*, and R*Co/ar*, were defined as the ratios of methyl/methylene, aromatic/aliphatic, carboxyl/aromatic, and carbonyl/aromatic, respectively. The carbonyl groups include ester, carboxyl, and other carbonyl group such as ketone (Ohki et al., 1999).

Table 2. Parameters based on FTIR spectra of the raw and the upgraded coals

Coal	RCH_3/CH_2	Rar/al	RCOOH/ar	RCO/ar
Raw Berau Coal	0.22	1.23	0.14	0.31
Upgraded Berau coal	0.29	3.48	0.17	0.27
Raw Tabang Coal	0.64	1.99	0.10	0.31
Upgraded Tabang Coal	0.81	2.17	0.10	0.27
Raw Samaranggau Coal	0.12	3.00	0.02	0.32
Upgraded Samaranggau Coal	0.36	3.51	0.02	0.31

RCH_3/CH_2 = 2965 cm^{-1} band/2920 cm^{-1} band
Rar/al = 1615 cm^{-1} band/(total of 2965-2850 cm^{-1}, five bands)
RCOOH/ar = 1710 cm^{-1} band/1615 cm^{-1} band
RCO/ar = (total of 1770-1655 cm^{-1}, four bands)/1615 cm^{-1} band

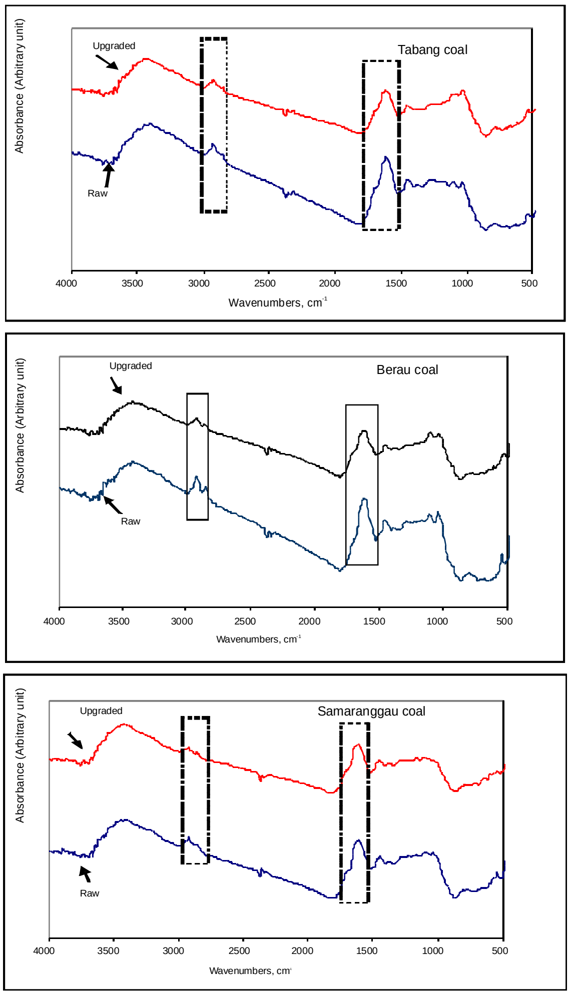

Figure 3. FTIR spectra for the raw and upgraded coals

The value of RCH_3/CH_2 in the zone of aliphatic hydrogen of the upgraded low rank coals increased. It is ruled that when low rank coals are upgraded, increase in methyl/methylene ratio generally takes place (Mahidin, 2003). The decrease of hidrogen in the upgraded low rank coal was due to the decrease of moisture content which consisted of hydrogen and oxygen atoms. While the increase of RCH_3/CH_2 was caused by the released/reformed of the hydrogen in the C-H functional group (aliphatic hydrogen) due to the heat (Painter et al, 1985) during the UBC process the CH_3 of the upgraded low rank coals were increased. These were shown in the increasing of the 2965 cm^{-1} bands compared to those of the raw coals.

The value of *Rar/al,* which related to the aromaticity of coal, the upgraded low rank coals increased, R*COOH/ar* of the upgraded Berau coal increased and upgraded Tabang and Samaranggau coals were the same. The value of R*CO/ar* for all of the upgraded low rank coals slightly decreased. As mentioned above, this phenomenon is consistent with the results of the oxygen content in the ultimate analysis.

Petrography Analysis

Based on coal petrography analysis to Tabang coal, there was no significant quantity and textural differences of maceral in the raw and the upgraded coals, but the mean vitrinite reflectance was slightly higher in the upgraded coal compared to that in the raw coal. Although vitrinite reflectance differences were small but it was significant especially if assessed in terms of the standard deviations. It should be remembered, however, that vitrinite reflectance values were obtained for low rank coals which were strongly affected by the atmospheric conditions. A small number of circular voids were found in the upgraded coal, that holes might have formed during the evolution of moisture in the steam phase.

Vitrinite was dominant in the samples of both the raw and the upgraded coals with the average content of 82.6%, followed by inertinite 9.2% and liptinite 7.1%. Mineral matter content, mainly silica and clay minerals was only 1.1% in the sample. Vitrinite mostly occured as telovitrinite (predominantly textinite texto-ulminite, eu-ulminite and lesser telocollinite) and detrovitrinite (attrinite, densinite and desmocollinite) with gelovitrinite as a minor component. Inertinite, which was generally associated with vitrinite comprised predominantly semifusinite (occured as layers, lenses or isolated fragments), sclerotinite (consisting of unilocular, bilocular teleutospores and sclerotia) and inertodetrinite. Liptinite comprised predominantly resinite (occured as discrete bodies, layers and lenses), suberinite (commonly occured in association with corpogelinite), liptodetrinite and cutinite with minor sporinite.

Mean vitrinite reflectance of the raw coal was 0.38% (0.29 – 0.44%) with 0.039 standard deviation. The upgraded coal had slightly higher vitrinite reflectance with a mean value of 0.45% (0.35 – 0.60%) with 0.053 standard deviation. The same trend was also indicated by specific energy which increased from air dried raw coal (5048 kcal/kg, adb) through Upgraded coal (6310 kcal/kg, adb).

Strong greenish yellow fluorescence of oil cut and exsudatinite were present in the raw and the upgraded coals with no significant difference of fluorescence colour. This was assumed to indicate that there was no chemical degradation and no loss of volatile hydrocarbons during the UBC process which was needed to change the fluorescence properties. Chemical reaction did not occur during the process, therefore none internal coal texture and structure changes were found. This was true because the UBC process was operated under low temperature and pressure conditions.

Specific Surface Area

Coal upgrading process is expected to reduce the specific surface area of the raw coal because during UBC process, the additional of oil to prevent the moisture reabsorption, may coat the surface permanently. As can be seen in Table.3, the specific surface area of upgraded coal is lower than that of raw coal. The decreases of the surface area may also be explained by plugging of pore structure and shrinkage by adding residual oil. The specific surface area of the coals decreased by increasing the coal particle size (see Table 3).

Briquettability

To study the briquettability of UBC, drop shutter and compressive strength were tested. The result of drop shutter test is shown in Table 4. It can be seen that the fraction size of -37.5+25 mm, which is the largest fraction in this test, produces the highest mass fraction for all of the low rank upgraded coals. This result indicates that the upgraded low rank coals briquette is quite strong, not easy to be broken caused by friction one to each other.

Table 3. The BET surface area of the raw and upgraded coals

Coals	Specific surface area, m^2/gr		
	Coal size		
	200 mesh	60+200 mesh	60 mesh
Raw Berau Coal	6.65	5.78	5.12
Upgraded Berau coal	5.01	4.78	3.79
Raw Tabang Coal	6.68	6.45	4.4
Upgraded Tabang Coal	5.58	5.03	3.84
Raw Samaranggau Coal	7.27	5.75	4.84
Upgraded Samaranggau Coal	6.59	4.58	3.63

Table 4. Upgraded Low Rank Coals Briquetting Test Results

	Upgraded Low Rank Coals		
Fraction (mm)	Berau	Tabang	Samaranggau
-37.5+25	73.9	63.2	74.40
-25+19	3.54	14.2	10.91
-19+12.5	2.06	4.6	3.75
-12.5+6.3	3.73	4.4	4.35
-6.3+3.35	1.59	2.0	1.55
-3.35	15.1	11.6	5.00
Comp. strength kg/cm^2 (averages)	62.7	40.9	65.19

CONCLUSIONS

1. UBC process reduces moisture content of the coals and automatically increases the calorific value of the coals.

2. The ash contents of the upgraded low rank coals are slightly higher than that of the raw coal, except for Tabang coal. On the other hand, the volatile matter of the upgraded low rank coals increases, due to the kerosene or oil that coats on the surface of coal.

3. The carbon content of the upgraded low rank coals significantly increases with increasing fixed carbon. The removal of water from coal causes the decrease of hydrogen and oxygen contents in the upgraded coals. While total sulfur and nitrogen contents slightly change.

4. The functional group of C-H and C=O of the upgraded low rank coals are slightly less than those of the raw coals.

5. Based on the coal petrography analysis, there is no significant quantity and textural differences of maceral in the raw and the upgraded low rank coals, but the mean vitrinite reflectance is slightly higher in upgraded coal than that in the raw coal.

6. The specific surface areas of the upgraded low rank coals are lower than those of the raw coals. The higher the coal particle size, the lower the specific surface area is.

7. The drop shutter test results of the upgraded low rank coals briquette show that the fraction size of -37.5+25 mm, which is the largest fraction in this test produces the highest mass fraction for all of the upgraded low rank coals briquette.

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