CHEMICAL AND PHYSICAL PROPERTIES OF UPGRADED BROWN COAL

DATIN FATIA UMAR [1,2], HIROMOTO USUI [2], BUKIN DAULAY [1], IWAN RIJWAN [1] AND IKIN SODIKIN [1]

[1] Research and Development Centre for Mineral and Coal Technology
Jalan Jenderal Sudirman No. 623 Bandung

[2] Department of Chemical Science and Engineering, Kobe University
Rokkodai, Nada-ku, Kobe, JAPAN

ABSTRACT

Results of proximate analyses indicate that inherent moisture of the upgraded coals decrease significantly compared to that of the raw coals. Hence, the calorific value of the upgraded coals increases. The ash content of the upgraded coals did not change obviously due to the UBC process which was conducted at low temperature. However, the volatile matter content increase slightly due to the residue plugs over coal pores to prevent re-absorb of moisture. From ultimate analyses, carbon content of the upgraded coals increases, whereas the hydrogen and oxygen contents decrease. The UBC process hardly effects to the sulfur and nitrogen contents. The equilibrium moisture of the upgraded coals was determined by using ASTM Standard method, most of them were less than 9%. The functional group of C-H and C=O of the upgraded coals were slightly less than that of the raw coals. The aromaticity of coal, all of the upgraded coals was increase. The petrography of both the raw and the upgraded coals indicates that the mean vitrinite reflectance was slightly higher in the upgraded coal compared to that of 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 coals due to the plugging of pore structure and shrinkage by residual oil addition. The briquettability of Upgraded coal briquette according to drop shatter test and compressive strength indicates good characteristics of briquette.

Keywords: low rank coal, UBC (Upgraded Brown Coal), raw coal, inherent moisture

1. INTRODUCTION

Coal deposits are widely distributed throughout Indonesian archipelago performing total resources of to 57.8 billion tons. Its reserve production ratio (R/P) 76 years. Classified as lignite of 58%, sub-bituminous of 27%, bituminous of 14% and small amount (less than 1%) of anthracite. Lignite and sub-bituminous coal have high moisture content, high volatile matter content, low calorific value and high oxygen content. Referring to these properties, it is uneconomic to carry out over long distances transportation. Therefore, precaution has to be done to prevent spontaneous combustion from the coal which dried partially. One questions is whether a low rank coal (LRC) can be upgraded in the form of energy, which can be transported economically and storage safely.

Several methods of dewatering and upgrading processes have been studied since 1920s (Suwono and Hamdani, 1999) to reduce moisture content and produce coal with higher calorific value and lower transportation cost (Suwono and Hamdani, 1999). Among them, UBC (upgraded brown coal) process developed by Kobe Steel Ltd., Japan will be applicable (Deguchi, et. al., 2002). The process upgrades a LRC and reduces moisture content to be equal to high rank coal. Therefore, it is expected to decrease the susceptibility to spontaneous combustion.

Process condition 0.35 MPa, was milder compared to that of other upgrading methods. Addition of 1% residual oil was very important to prevent the moisture re-absorption. However long distance transportation, UBC powder needs to be briquetted using a double roll briquetting machine without the binder addition.

The objective of this study is to provide general knowledge that related to UBC process as basic information on both raw and upgraded coals characteristics in order to establish the advantages of the process from the technical view of points. Information of this problem is very valuable to help make decisions for the process that is needed in selecting process condition in terms of increasing process efficiency.

This paper discusses the characteristics of raw and upgraded LRC. Two samples of Berau and Taban, East Kalimantan and one sample coming of Samaranggau, South Kalimantan have been processed to their the changes due to UBC process.

2. EXPERIMENT

The raw coals from 3 coal mining areas in Indonesia, had been processed in UBC pilot plant at Palimanan, Cirebon. The process consisted of five main sections, namely coal preparation, slurry dewatering, coal-oil separation, oil recovery and coal briquetting (Daulay, et al, 2003). Raw and upgraded coals had been analyzed to study the changes of chemical and physical characteristics prior and after UBC process. Proximate and ultimate analyses including equilibrium moisture and calorific value in air dried basis (adb) had been conducted according to the ASTM Standard.

A functional group analysis had been performed to investigate the change of quality due to the upgrading by UBC process. A functional group analysis had been conducted by a Fourier transform infrared HORIBA FT-720 spectrometer (FTIR) with the resolution of 1 cm^{-1}. Sampling was carried out by the KBr pellet method and measurements were conducted in spectra selected range of 3000-2700 cm^{-1} and 1800-1500 cm^{-1}. That ranges were assigned to the aliphatic hydrogen (C-H functional group) and oxygen containing structures (C=O functional group), respectively (Ma, Hill, and Heng, 1989).

Coal petrography analysis had been carried out by polarization microscope MPM-200. Petrography characteristics of coal were conduction in terms of two essential independent concepts, i.e., coal type and coal rank. Coal type refers to the nature of the organic matter within in coal and coal rank refers to the stage of coalification that has been reached by the organic matter. Therefore, petrographic study of coal is based on the morphology, color, particle size, reflectance and anisotropy of the macerals in reflected white light and auto fluorescence of the constituents under blue-ultraviolet radiation. This research intends to determine if the upgraded coal can petrographically be distinguished from the raw coal.

To study physical properties of both raw and upgraded coals, the specific surface area had been measured by BET (Brunauer Emmet Teller) method using CO_2 at 25°C and SHIBATA APPSA-100 based on the amount of nitrogen absorbed, by the surface of solid particles. The nitrogen flow rate was 150 ml/min. Prior the test, coal samples had been dried in nitrogen atmosphere at 180°C. Drop shatter test and uniaxial compressive strength of upgraded briquette had also been studied.

3. RESULTS AND DISCUSSION

3.1 Proximate and Ultimate Analyses

Results of proximate and ultimate analyses which include equilibrium moisture and calorific value of raw and upgraded coals are shown in Table 1. All raw coals indicate high moisture content and low calorific value. Low rank coals contain appreciable amounts of oxygen in the form of carboxylic acids and salts of carboxylic acids (Deguchi, 2002). This fact contributes to the low heating value due to their affinity to moisture. All upgraded coals show a decrease in moisture content and an increase in calorific value compared to that of raw coals. It means that most of moisture content in the raw coal can be removed by UBC process. The increase of calorific value of the upgraded coal promises the utilization of this coal can be enhanced. Figure 1 and 2 illustrate the decrease of moisture contents and increase of calorific value within upgraded coals compared to that of raw coals, respectively. Based on the equilibrium moisture (EM) content, most of the upgraded coals have EM content less than 9%.

Ash content of all of upgraded coals was slightly higher compared to that of the raw coals. The increase of ash content of the upgraded coals related to residual oil addition that plugs coal pores. It was identified as ash content in the coals. However, in general, the increase of ash content in the upgraded coals compared to that of the raw coals

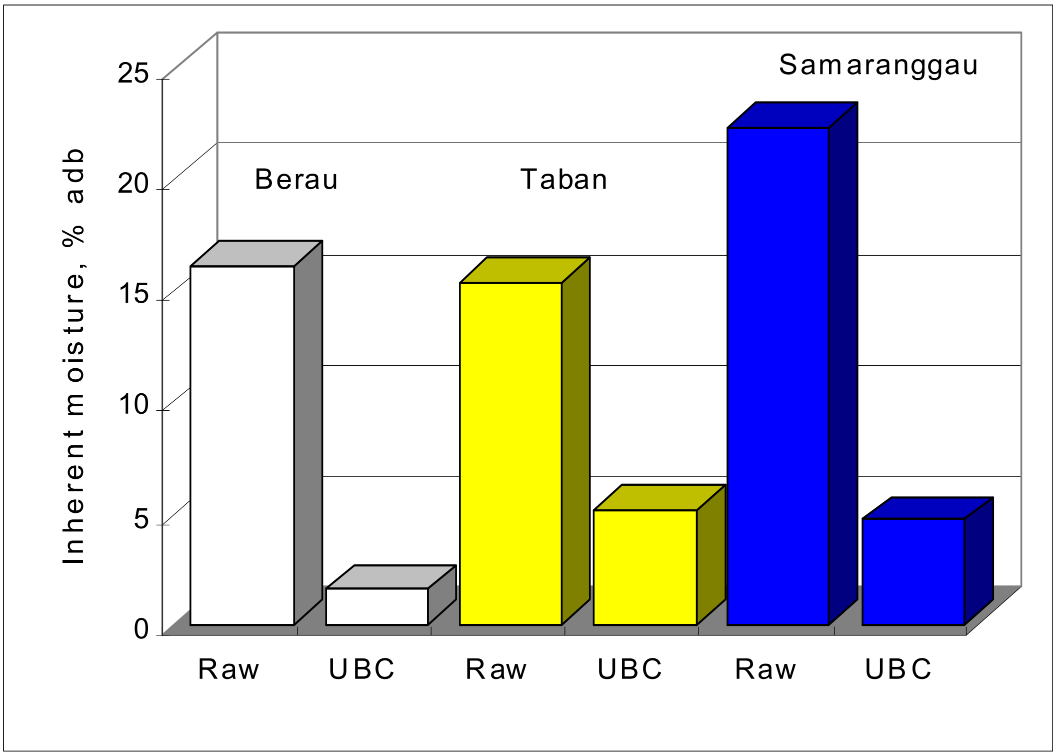

Figure 1. Moisture content of raw and upgraded coals

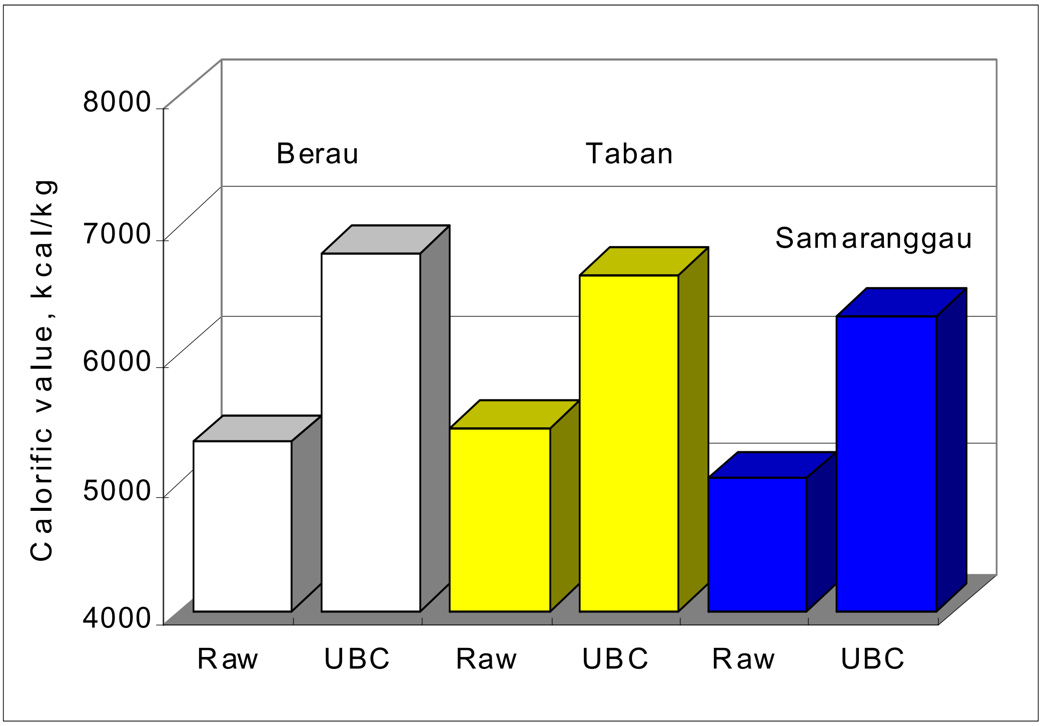

Figure 2. Calorific value of raw and upgraded coals

is insignificant. On the other hand, the volatile matter of all upgraded coals increased. This was caused by kerosene or residual oil that coated the upgraded coal surface. Kerosene or oil was detected as volatile matter when coal was burned. The increase of fixed carbon certainly related to reduction of moisture content.

Based on ultimate analyses, carbon content of upgraded coals significantly increases related to increase of fixed carbon. The increase of carbon content will improve coal combustibility. It means that coal utilization as a fuel will also be enhanced. Water removal 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 upgraded coals probably related to the residual oil which was added in the process to prevent the re absorb of moisture.

3.2 Functional Group Analysis

The FTIR spectrum for 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. Zone of aliphatic hydrogen corresponds to asymmetric methyl ($-CH_3$) and methylene ($-CH_2-$), methane (C-H) and symmetric methyl ($-CH_3$) and methylene ($-CH_2-$). It was observed in 2965, 2920, 2895, 2870 and 2850 cm^{-1}, respectively. Moreover 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 carboxylate groups (COO-) stretching at 1770, 1700, 1655,1615, 1580, 1560 and 1540 cm^{-1}, respectively.

Compared to the peak of the C-H and C=O group of raw and upgraded coals, the peak of upgraded coals is slightly lower than that of raw coals. Hydrogen and oxygen are almost equal or slightly less than those of raw coals as shown in Table 1. It is caused is the UBC process had been conducted under mild condition.

The following parameters are defined as the ratio of deconvoluted peak area, R*CH3/CH2*, R*ar/al*, R*COOH/ar*, and R*Co/ar* are defined as ratios of methyl/methylene, aromatic/aliphatic, carboxyl/aromatic, and carbonyl/aromatic, respectively. Carbonyl groups include ester, carboxyl, and other carbonyl group such as ketone (Ma, S., Hill, J. O., and Heng, S. J., 1989).

Rations are as follows :
RCH_3/CH_2= 2965 cm^{-1} band/2920 cm^{-1} band
R*ar/al*= 1615 cm^{-1} band/(total of 2965-2850 cm^{-1}, five bands)
R*COOH/ar*= 1710 cm^{-1} band/1615cm^{-1} band
R*CO/ar* = (total of 1770-1655 cm^{-1}, four bands)/ 1615 cm^{-1} band

FTIR parameters of raw and upgraded coals before and after UBC processed are summarized in Table 2. RCH_3/CH, value of the upgraded coals increased. It is show that when low rank coals were upgraded. Methyl/methylene ratio incolase generally occous.

Rar/al which related to the aromaticity of coal, all of the upgraded coals value increased, R*COOH/ar* of the upgraded Berau coal was also increased. The upgraded Taban and Samaranggau coals were also equal. The value of R*CO/ar* for all of upgraded coals is slightly decreased. This phenomenon is consistent with the results which were obtained from ultimate analysis of the oxygen content.

3.3 Petrography Analysis

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

Vitrinite is dominant in samples both raw and upgraded coals. The average content is 82.6%, followed by inertinite (9.2%) and liptinite (7.1%). Mineral matter content, mainly silica and clay minerals, is only 1.1% in the sample. Vitrinite mostly occurs as telovitrinite (predominantly textinite texto-ulminite, eu-ulminite and lesser telocollinite) and detrovitrinite (attrinite, densinite and desmocollinite) with gelovitrinite a minor component. Inertinite, generally associated with vitrinite, comprises predominantly semifusinite

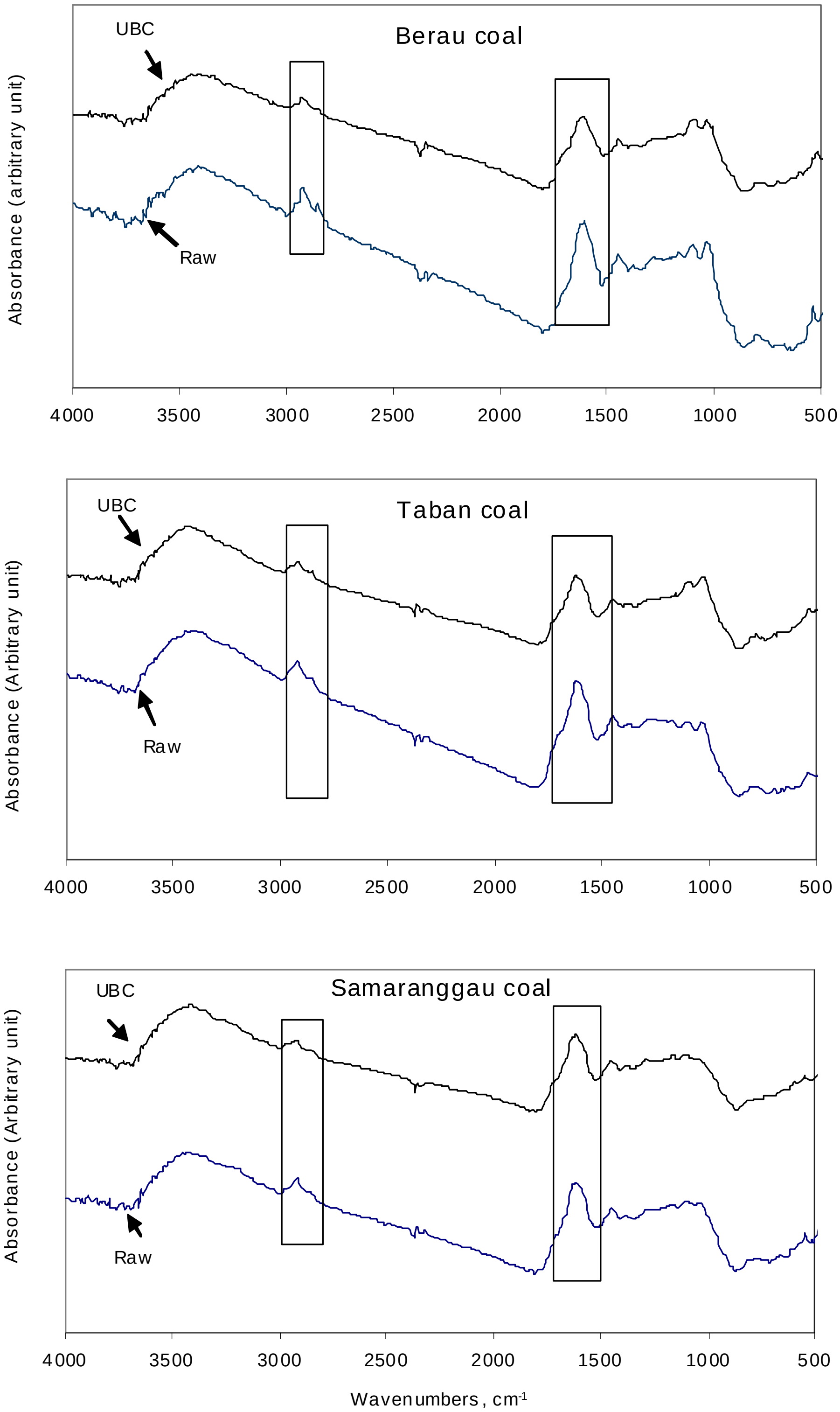

Figure 3. FTIR spectra for raw and upgraded coals

Table 1. Analyses results of raw and upgraded coals

Coals	Berau		Taban		Samaranggau	
Parameter, air dried basis	Raw	UBC	Raw	UBC	Raw	UBC
Inherent Moisture, %	16.13	4.81	15.35	5.49	22.33	4.65
Ash, %	6.36	6.81	4.42	5.39	2.15	2.61
Volatile matter, %	37.20	46.69	40.68	46.13	38.05	47.67
Fixed carbon, %	40.31	45.38	39.55	46.57	37.47	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	65.91	57.55	67.61	53.6	64.43
Hydrogen, %	6.57	6.28	6.06	5.17	6.19	5.30
Nitrogen, %	1.60	1.03	0.77	0.82	0.69	0.86
Oxygen, %	28.34	23.18	31.06	20.78	37.3	26.64
Calorific value , kcal/kg	5324	6274	5431	6625	5048	6310

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

Coals	RCH_3/CH_2	R*ar/al*	R*COOH/ar*	RCO/ar
Raw Berau	0.22	1.23	0.14	0.31
UBC Berau	0.29	3.48	0.17	0.27
Raw Taban	0.64	1.99	0.10	0.31
UBC Taban	0.81	2.17	0.10	0.27
Raw Samaranggau	0.12	3.00	0.02	0.32
UBC Samarnggau	0.36	3.51	0.02	0.31

(occurs as layers, lenses or isolated fragments), sclerotinite (consisting of unilocular and bilocular teleutospores and sclerotia) and inertodetrinite. Liptinite comprises predominantly resinite (occurs as discrete bodies, layers and lenses), suberinite (commonly occurs in association with corpogelinite), liptodetrinite and cutinite with minor sporinite.

Mean vitrinite reflectance of raw coal is 0.38% (0.29 – 0.44%) with 0.039 standard deviation. The upgraded coal has slightly higher vitrinite reflectance with a mean value of 0.45% (0.35 – 0.60%) with 0.053 standard deviation. The same trend is also indicated by specific energy which increase from raw coal air dried (5324 kcal/kg, adb) through upgraded coal (6274 kcal/kg, adb).

Strong greenish yellow fluorescence of oil cut and exsudatinite is present in raw and upgraded coals with no significant difference of fluorescence color. It is assumed to indicate that no chemical degradation and no loss of volatile hydrocarbons during the UBC process. Chemical reaction did not occur during process, therefore no internal coal texture and structure changes. This is true because the UBC process was operated under low temperature and pressure conditions.

3.4 Specific Surface Area

Coal upgrading process expects that specific surface area of the upgraded coal is lower than that of raw coal. Because during UBC process, the additional oil to prevent re-absorb of moisture can be removed permanently. As can be seen in Table 3, the specific surface area of upgraded coals is lower than that of raw coals. Decreases of surface area may also be explained by plugging of pore structure and shrinkage by added residual oil. Coal particle size hardly affects the specific surface area.

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

Coals Coal size	Specific surface area, m^2/gr		
	200 mesh	60+200 mesh	60 mesh
Raw Berau	6.65	5.78	5.12
UBC Berau	5.01	4.78	3.79
Raw Taban	6.68	6.45	4.40
UBC Taban	5.58	5.03	3.84
Raw Samaranggau	7.27	5.75	4.84
UBC Samaranggau	6.59	4.58	3.63

3.5 Briquettability

To study the briquettability of the upgraded coals, drop shatter test and uniaxial compressive strength had been conducted. Result of drop shatter test is shown in Table 4. The fraction size of -37.5+25 mm, which was the highest fraction in this test, produces the highest mass fraction for all of upgraded coals This result indicates that the upgraded coal briquettes are quite strong and are not easy to be broken due to friction one to another as reflected by high compressive strength.

Table 4. Test results of drop shatter

Fraction (mm)	Coals, mass fraction weight %		
	Berau	Taban	Samaranggau
-12.5	73.90	63.20	74.40
-6	3.54	14.20	10.91
-6.5	2.06	4.60	3.75
-6.2	3.73	4.40	4.35
-2.95	1.59	20.00	1.55
-3.35	15.10	11.60	5.00
Comp. strength kg/cm^2 (averages)	62.70	40.90	65.19

CONCLUSIONS

Experimental study of this work has given the following conclusions:

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

2. Ash content of upgraded coals slightly increase compare to that of raw coals. Volatile matter of the upgraded coals also increases.

3. Carbon content of upgraded coals significantly increase as the increase of fixed carbon. Water removal 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 upgraded coals is slightly less than that of the raw coals.

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

6. Specific surface area of upgraded coals is lower than that of raw coals. Coal particle size hardly affects surface area.

7. Drop shatter test results of upgraded coals briquette show that the fraction size of -37.5+25

mm, the highest fraction in this test, produces the highest mass fraction for all of the upgraded coals. It is reflected by the high compressive strength.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the JSPS Ronpaku fellowship program, Dr. Balia, - Head of R & D Centre for Mineral and Coal Technology (*tek*MIRA) for providing opportunity to conduct this research. Thanks also for the staffs of coal laboratory of *tek*MIRA Dr. Mustafa, Mr. Indartono, Mr. Nguyen and Mr. Ikeda, for their full assistance during laboratory works.

REFERENCES

Daulay B., Salim, A. D., Otaka, Y., Deguchi, T., Makino, E., Rijwan, I., and Umar, D. F., 2003. *Proc. of The 4th International Conference and Exhibition on Coal for Power and Economic Development, Coal-Tech 2003*, Indonesia.

Deguchi, T., Shigehisa, T. and Shimasaki, K., 1999. *Proc. International Conference on Clean and Efficient Coal Technology in Power Generation*, Indonesia, pp.176-180.

Deguchi, T., Shigehisa, T., Makino, E. and Otaka, Y., 2002. *Proc. International Conference and Exhibition on Clean and Efficient Coal Technology in Power Generation, Coal-Tech 2002*, Indonesia.

Ma, S., Hill, J. O., and Heng, S. J., 1989. *Journal of Thermal Analysis,* 35, pp. 1985 1996.

Ohki, A., Xiao, F.X., Nakajima, T., Itahara, T. and Maeda, S., 1999. *Coal Preparation,* 21, pp 23-24.

Suwono, A. and Hamdani, 1999. *Coal Preparation,* 21, pp. 141-153.