# The Use of Mixed Oils Coating for Upgrading Brown Coal

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# The Use of mixed oil coating for Upgrading Borneo Brown Coal

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## Abstract

Indonesia (Borneo) is an important producers of coal in the world. Unfortunately, the type of coal is mostly in the low-rank coal which is called brown coal. This study was aimed to evaluate influence of mixed oil coating to upgrade Borneo brown coal. The additive material used to upgrade brown coal is mixed oil such as used cooking oil and waste lubricant in various ratios. Coal samples were pulverized in range size 10 - 20 mesh, mixed, dried, and coated. Evaluation of heat thermal values and water readsorption characteristics of original and coated coal have been done. Compared to original coal, heat thermal values of coated coals were significantly improved. The result showed that coated coals have a resistance to water re-adsorption. In addition, effect of mixed oil coating highly significant to have high isothermal oxidation due to suppression of active functional groups to oxygen in air. The study results also showed that increasing caloric value and lowering water content significantly can be achieved by these mixed oil

Key Words : brown coal, coating, mixed oil, thermal heat value

## 1. Introduction

Indonesia (Kalimantan) is the five largest in the world as producers of coal. Unfortunately, the type of coal is mostly in the low-rank coal which is called brown coal [1]. Globally, low rank coal constitute more than 50 % or more than 90% of the Indonesia's Borneo Brown coals coal reserve (BCs) [2, 3]. The price of low rank coal much lower compare to high rank coal then BCs attracts attention around the world. However, because of their lower heating values (5100 – 6100 kcal/kg), and high moisture content (25 – 60%) [4] the utilization of BCs are limited [2]. High moistures content is a big problem because they influence on high transportation cost, fuel efficiency, and combustion energy products [5]. Government of Indonesia encourages some research in order to enhance added value of both low and medium rank coal [2]. Afterward, improve heating value and decreasing moisture content of low rank coal in order to increase the application of BCs become an interesting study. A characteristic of low rank coal, they have high tendency toward spontaneous combustion [6]. Upgrading of BCs is one promising way to meet these requirements.

Now days, Upgrading of BCs by dewatering technology is the favorite techniques [7]. This method has significantly improved heating value and reduced the moisture content of brown coal. Therefore it can reduce sensitivity of coal toward spontaneous combustion [8]. Unfortunately, after drying, an upgraded coal may have more pore than original, which make their moisture re-adsorption more sensitive [9]. High moisture content not only relate to spontaneous combustion more potential but also potential to make the heating value become lower [6, 10]. The investigated of upgrading BCs by adding some fresh heavy oil have been done by several studies but in terms of operational these methods is expensive because of raw material cost [3, 4] therefore, the use of alternative oil is required.

During BCs storage, an exothermic reaction spontaneous combustion is highly potential occurred where oxidation of BCs at low temperatures (<200°C) may be occur. This is one of the biggest problem that important to be discussed regarding the safety issue [11]. The spontaneous combustion reaction cause serious environmental problems in terms of emitted greenhouse gas such as carbon monoxide (CO) and carbon dioxide ($CO_2$) [12]. To evaluated kinetics model of reaction producing CO and $CO_2$ from LTO of coal, and the factors influencing the emission of carbon gaseous in the exit gases both oxygen consumption and production rate were the main parameters measured for the evaluation of study [13, 14]. An alternative upgrading methods strongly needed that can achieve the purpose mentioned above .

In this study, Indonesian BCs was upgraded by adding used cooking oil, and waste lubricant in various ratios. It is expected that said material would be useful to increase the heat value of upgraded BCs [15, 16]. In addition mixed materials are stored at room temperature in the liquid state and have ability to resistance on moisture [17]. Because of recycled oil, these materials very economist compared to fresh oil so the utilization of them for BCs upgradation additive should be at lower operational cost relatively. Furthermore, the present study is focused on the observation impact of mixed recycled oil for BCs upgradation on characteristics of the upgraded BCs. The measured thermal heat value of mixed oil coating is 60.75 x $10^3$ kJ/kg, so it is expected could be effectively improve the heating value of upgraded BCs.

## 2. Material and Method

Samples of Indonesian (Borneo) brown coal was used as a raw coal. The raw coal were cleaned from impurity in order to obtain good coating, then they were stored at room temperature with sufficient lighting. Representative samples were crushed and screening to obtain the required size 10 - 20 mesh ranges for analyses and experimentations. Each 50 g sample was mixed with the used cooking oil (UCO) and waste lubricant oil (WLO) at various ratios. The waste lubricant oils were obtained and collected from oil service station while UCO can be found in huge quantities such as restaurants, food courts and cafeterias. UCO and WLO were the additive agent used to stabilize the BCs.

Heating value and proximate analysis raw and upgraded coals were conducted by using Bomb Calorimeter Parr 6200 ASTM D 5865-10a method and ASTM D3172-07a using TGA701 thermogravimetric analyzer respectively. The properties of BCs and additive are given in Table 1. The surface areas of the coal samples were examined using Brunauer-Emmett-Teller (BET) method [18]. Prior to BET surface area analysis, the samples were heated to 110°C for 12 h in a $N_2$ environment. BET analysis starts by cooling down the sample to ~ 77 K, followed by nitrogen injection under various pressure to determine $N_2$ displacement for specific are calculation. [19]

## 3. Result and Discussion

### 3.1 Characteristic of Raw and Upgraded Coal

Proximate analysis and thermal heat values of raw and upgraded coals shown at Table 1. Moisture content of upgraded coal decreased up to 15 – 30 % (w/w). Originally raw coals had 17.75 % (w/w) moisture content. Not only decreasing moisture content of upgraded coal at the same time mixed oil can increase thermal heat values significantly. In the case of thermal heat value raw coal which originally has 4868 kcal/kg increase up to 6412 kcal/kg maximum with ratio waste lubricant: used cooking oil at 1 : 2 .

Table 1. Raw and treated coal proximate analysis and heating values

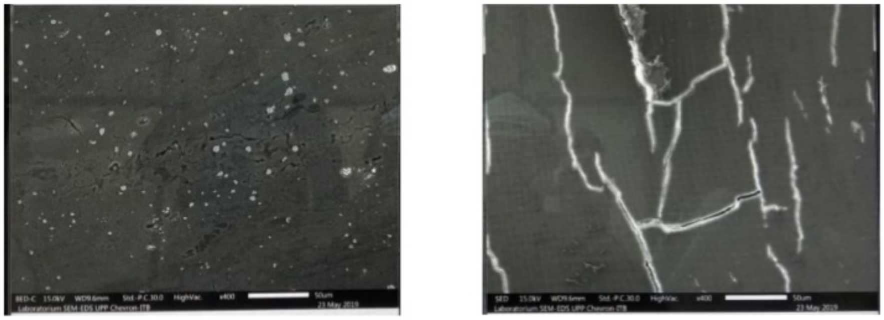
| Samples | Moisture % | HV Kcal/kg | Volatile % | Ash % | FC % |
|---|---|---|---|---|---|
| BCs (as received) | 17.75 | 4868 | 36.34 | 5.53 | 29.77 |
| UCO | 3.62 | 7283 | - | - | - |
| WLO | 5.96 | 6166 | - | - | - |
| BCs-mixed oil | | | | | |
| Ratio WLO:UOC (v/v) | | | | | |
| 1:0.25 | 4.2 | 5443 | | - | - |
| 1:0.75 | 3.9 | 5627 | | - | - |
| 1:1 | 3.7 | 5822 | | - | - |
| 1:1.5 | 3.61 | 6234 | | - | - |
| 1:2 | 3.74 | 6412 | | - | - |
| 1:3 | 4.1 | 6356 | | - | - |
| 1:4 | 4.75 | 6243 | | - | - |

In order to evaluate effect of coating on treated coal for surface area and pore volume some testing, using BET, was done as Table 2 shows. The BET's testing result shows that surface area in raw coal was 2.41 $m^2/g$ and after coating it reduced sharply up to 0.97 $m^2/g$. This potentially occurred due to the fact that the upgrading additive fulfill the pores and replace the hydrate water during the process of upgrading. In addition, a same trend, as compared to raw coals, in pore volumes after coating, particles also experience coating and clogging with each other.

Table 2. Original and treated coal surface area and pore volume

| Samples | Surface Area, BET ($m^2/g$) | Pore Volume ($cm^3/g$) |
|---|---|---|
| BCs (as received) | 2.41 | 0.0172 |
| BCs-mixed oil 10% (v/v) WLO: UCO 1:2 | 0.97 | 0.0079 |

Fig. 1(a) shows a macroscopic image of raw coal before coating and Fig. 1(b) shows the upgraded coal respectively. The shinning parts of the images indicate the additive oils coating occupy the pore of particles coal surface after drying step. On the other hand the oils coating UCO and WLO was spreaded on the coal surface very well. It means the upgraded coal should be well protected from the moisture re-adsorption and oxidation.



(a) (b)

**Fig. 1 (a) Macroscopic image of the raw coal (b) treated coal particles (10-20 mesh)**

### 3.2 Upgraded-Coal Coating Process

BCs was feed in to a secondary mill to have smaller particle and sorted by screen to have particle size in range 10 - 20 mesh. To have the moisture content all samples similar around 10% pulverized coal were put in oven at 110°C for 70 min. Drying also provide more pore so that mixed oil can coat the upgraded coal. Each 50 g sample was mixed with oil coating which heated until 200ºC and continuously stirred at constant speed 100 rpm in order to intimate contact the surface coating of coal particles. The upgrading coal separate from oil coating using a vacuum filter. A briefly step process of upgraded coal manufacturing process was description in Kinoshita et al. [20]

### 3.3 Characteristics Water re-adsorption

Water re-adsorption characteristics of BCs before and after coating were examined by equilibrium moisture method. Standard Test Method for Equilibrium Moisture of Coal at 97 Percent Relative Humidity and 30°C, ASTM D-1412-02a, was used [4]. This goal was conducted where 50 g coal was put into the glassware. It was stirred in water bath for 12 hours and then store in humidity chamber for 72 hours at 97% relative humidity and 30°C as standard procedure explained by Khan et.al [5, 21]. Furthermore, the moisture content of dried and upgraded were determined by proximate analysis. The results of experiments can be seen at Figures 2.

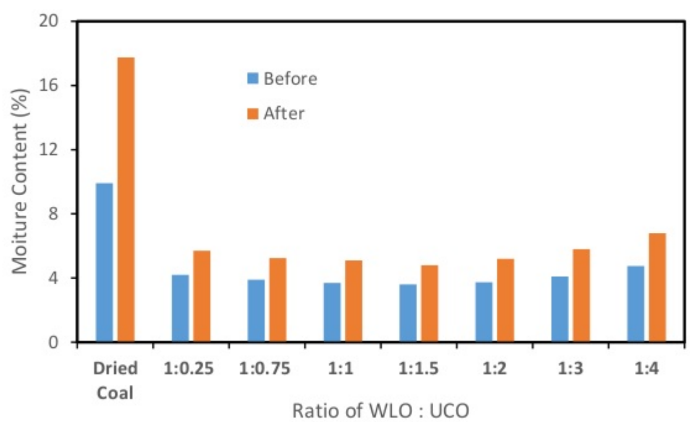



**Figure 2. Water re-adsorption characteristics original and treated coal**

### 3.4 Evaluation of degree of spontaneous combustion

Spontaneous combustion reaction is one of the important problems of coal. All coals oxidize when they start to be exposed to air, especially during and after mining [22]. This tends to be more of a problem in lower rank coals like lignite. One of parameter that can be used to assessing coal characteristic is crossing point temperature (CPT) or relative ignition temperature [5, 22]. It is the temperature at which the coal temperature begins to exceed the surrounding temperature. Reported that coal with a CPT of 120–140°C and a moisture content of more than 5% is highly susceptible to spontaneous combustion [22]. To evaluate the self-combustion characteristics of raw and upgraded coal, CPT was assessed. For this purpose, the 50 g coal sample is heated in a reaction tube in a furnace for 2 hours at rising temperature; with nitrogen passing through it at 80 ml/min rate till the coal temperature crosses the furnace temperature. The point at which the temperature of the furnace started to increase as compared to the coal temperature was considered as the CPT. Figure 3 shows that CPT of treated coal was improved significantly. A detailed description of CPT measuring apparatus is described in many literatures.

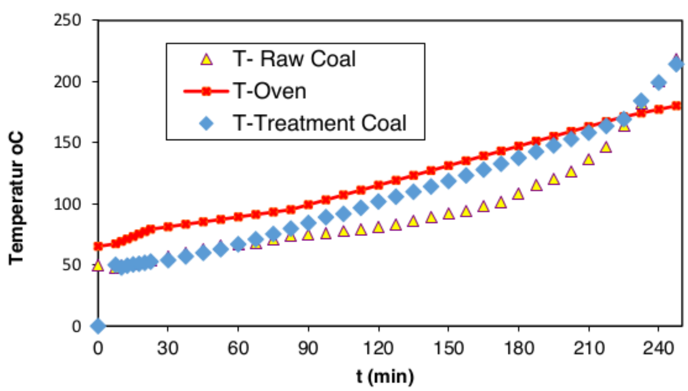



**Figure 3. CPT measurements**

### 4. Conclusion

In this report, low rank coal from Kalimantan (Borneo) Indonesia was upgraded by addition of waste lubricant oil and used cooking oil in various ratio. Characteristics of upgraded coals such as thermal heat values, spontaneous combustion, water re-adsorption and isothermal oxidation were evaluated. This study shows that mixed oil in ratio waste lubricant to used cooking oil 1: 2 has strong potential to improve low rank coal not only for higher heat calorific values but also for lower water re-adsorption, and finally for a medium tendency to spontaneous combustion.

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## References

[1] Heri Heriyanto, W. Ernayati, M. Yulvianti and Sutijan, "The Influence of Kerosene to Increase the Quality of Coal from Bayah South Banten Using UBC Methode," *World Chemical Engineering Journal,* vol. 1, no. 1, pp. 6-10, 2015.

[2] G. Gushka, "The Low Rank Coal Policies In Indonesia," Directorate General Of Mineral And Coal, Tokyo, 2013.

[3] M. Z. Khan, D. H. Chun, a. S. D. K. Jiho Yoo, a. H. K. C. J. L. Young Joon Rhim, S. Lee and A. Rifellaa, "Evaluation of the effect of a palm acid oil coating on upgrading low rank coal," *Royal Society of Chemistry Advanced,* vol. 5, no. 1, p. 63955–63963, 2015.

[4] M. Z. Khan, D. H. Chun, Y. . J. Rhim, S. D. Kim, J. Yoo, H. Choi, J. H. Lim, W. T. Jo and S. Lee, "Upgrading Indonesian Low-Rank Coal with an Oil Coating," *International Journal of Coal Preparation and Utilization,* p. 251–261, 2016.

[5] M. Z. Khan and D. H. Chun, "Upgrading Indonesian Low-Rank Coal with an Oil Coating," *International Journal of Coal Preparation and Utilization,* vol. 36, no. 5, p. 251–261, 2015.

[6] C. E. J. Center, K. Steel and Ltd, "UBC (Upgraded Brown Coal) Process Development," *Kobe Steel Engineering Reports,* pp. 77-78, 2003.

[7] Y. Huang, Y. Wang, H. Zhou, Y. Gao, D. Xu and L. Bai, "Effects of Water Content and Particle Size on Yield and Reactivity of Lignite Chars Derived from Pyrolysis and Gasification," *Molecules,* vol. 23, no. 10, p. 2717, 2018.

[8] A. A. Vostrikov, D. Y. Dubov, M. Y. Sokol, A. V. Shishkin and O. N. Fedyaeva, "Brown coal gasification in combustion in supercritical water," *Journal of Engineering Thermophysics,* vol. 25, no. 1, pp. 55-66, 2016.

[9] O. J and J. Willey and Sons, "Adsorption," *Ellis Herwood Series in Physical Chemistry,* 1994.

[10] "Study on Clean Coal Technology Project," The Institute of Energy Economics, Japan, Tokyo, 2012.

[11] J. D. N. Pone, K. A. Hein, G. B. Stracher, H. J. Annegarn, R. B. Finkleman, D. R. Blake, J. K. McCormack and Paul, "The spontaneous combustion of coal and its by-products in the Witbank and Sasolburg," *Coal Geology,* 2007.

[12] Indocement, "Proses UBC Batubara dengan bantuan minyak pelumas atau oli bekas," 2014.

[13] J. A. DeBarrl, M. Rostam-Abadi and S. A. Benson', "Combustion Properties of Illinois Coal-Char Blends," *Energy & Environmental Research* , 2000.

[14] L. Yuan and A. C. Smith, "CO and CO2 emissions from spontaneous heating of coal under different ventilation rates," Office of Mine Safety and Health Research, United State, 2011.

[15] O. L. E. and D. S. M. L. C. P, "Comparative study of calorific value of rapeseed, soybean, jatropha curcas and crambe biodiesel," in *International Conference on Renewable Energies and Power Quality (ICREPQ'13)* , Spain, 2013.

[16] D. V. R. Mamilla and D. L. N. R. G, "A review: Waste lubricating oil as an alternative fuel blended with diesel," *International Journal of Advanced Scientific Research,* vol. 1, no. 1, pp. 1-4, 2016.

[17] R. Permadi, L. Pulungan and Solihin, "Analisis Batubara Dalam Penentuan KualitasBatubara Untuk Pembakaran Bahan BAku Semen di PT. Indocemet Tunggal Prakarsa Tbk. Palimanan-Cirebon," Unisba , Bandung, 2015.

[18] G. N. Okolo, R. C. Everson, H. W. Neomagus, M. J. Roberts and R. Sakurovs, "Comparing the porosity and surface areas of coal as measured by gas adsorption, mercury intrusion and SAXS techniques," *Fuel,* vol. 141, pp. 293-304, 2015.

[19] D. r. n. B. Hellack, *SOP: Specific Surface Area Analysis by BET Theory,* 2016.

[20] S. Kinoshita, D. S. Yamamoto, T. Deguchi and T. Shigehisa, "Demonstration of Upgraded Brown Coal Process by 600 Tonnes/day Plant," P.T Upgraded Brown Coal Indonesia, Tokyo, 2010.

[21] S. Murata, K. Kidena, M. Hosokawa and M. Nomura, "Analysis of oxygen-functional groups in brown coals," *Fuel Processing Technology ,* pp. 231-243, 2000.

[22] D. L. L. Sloss, "Assessing and managing spontaneous combustion of Coal," IEA Clean Coal Centre, London, 2015.

[23] H. Choi, C. s. Thiruppathiraja, S. Kim, Y. Rhim, J. Lim and S. Lee, "Moisture readsorption and low temperature oxidation characteristics of upgraded low rank coal," *Fuel Processing Technology,* vol. 92, no. 10, pp. 2005-2010, 2011.

[24] K. Akiyama, H. Pak, Y. Ueki, R. Y. and I. Naruse, "Effect of Mg based addition to Upgraded Brown Coal on the Ash Deposition Behavior During Combustion," *MCS 7,* pp. 11-15, 2011.

[25] T. Sugita, "Low-rank Coal Upgrading Technology (UBC Process)," Kobe Steel Engineering Reports, Tokyo, 2003.

[26] G. Wang, J. Xie, S. Xue and H. Wang, "Laboratory Study On Low-TemperatureCoal Spontaneous Combustion in The Air Reduced Oxygen and Low Methane Consentration," *Technical Gazette,* vol. 22, no. 5, pp. 1319-1325, 2015.

[27] A. J. Al-Kofahi, "Waste Cooking Oil to Biodiesel Fuel," American University of Beirut, America, 2017.

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