IMPROVEMENT OF LOW RANK COAL PROPERTIES BY VARIOUS UPGRADING PROCESSES

DATIN F. UMAR and BUKIN DAULAY
R & D Centre for Mineral and Coal Technology
Jalan Jenderal Sudirman 623 Bandung 40211,
Ph. 022 6030483, fax. 022 6003373
e-mal: datinf@tekmira.esdm.go.id

ABSTRACT

A low rank coal from Banko, South Sumatera was used to study the properties improvement of the coal due to coal upgrading process. Various coal upgrading processes were conducted i.e. upgraded brown coal (UBC), hot water drying (HWD) and steam drying (SD). The UBC process was carried out in pilot scale with a capacity of 5 tons/day at temperature of 160°C and pressure of 0.35 MPa, while the HWD and SD processes were conducted in laboratory scale using autoclave at the temperature of 300ºC and pressure of about 12 MPa for 1 hour. The result indicates that the properties of Banko coals after the UBC, HWD and SD processes were improved. The calorific value of the upgraded coals was significantly increased in relevant to the decreasing of inherent moisture content and have better combustion characteristics than that of the raw coal. The highest calorific value can be achieved by SD process, followed by HWD and UBC processes.

Keywords: low rank coal, inherent moisture, calorific value

INTRODUCTION

Due to decreasing of oil reserves in some countries including Indonesia and the rise of the oil price in the world, the energy alternative to substitute of oil should be developed to support energy diversification policy. Among some energy alternatives resources, coal began to be reviewed due to its abundant resources of 104.77 billion tons in which 20.96 billion tons of them are reserves (Geological Agency in Kamandanu, 2010). The share of anthracite and bituminous coal only 0.3% and 14.3% respectively whereas most of them are classified as low rank coal.

According to the American Standard for Testing and Materials (ASTM), the low rank coals are lignite and sub bituminous coal which have:

- fixed carbon (dmmf, dry mineral matter free) <69%;
- volatile matter (dmmf, dry mineral matter free) >31%; and
- calorific value (mmf, moist mineral matter free) <11,000 kcal/kg.

LRC has also other characteristics as follows:

- high Total Moisture (20 – 40% as received) content;
- low Thermal Efficiency;
- low Ash Melting Temperature; and
- high Tendency for Spontaneous Combustion.

Based on Government Regulation (PP) No 45/2003 about non tax government income (PNBP) the quality of coal is divided into:

- low calorific value coal: calorific value < 5,100 kcal/kg
- medium calorific value coal: calorific value between 5,100 and 6,100 kcal/kg
- high calorific value coal: calorific value between 6,100 and 6,800 kcal/kg
- very high calorific value coal: calorific value > 6,800 kcal/kg

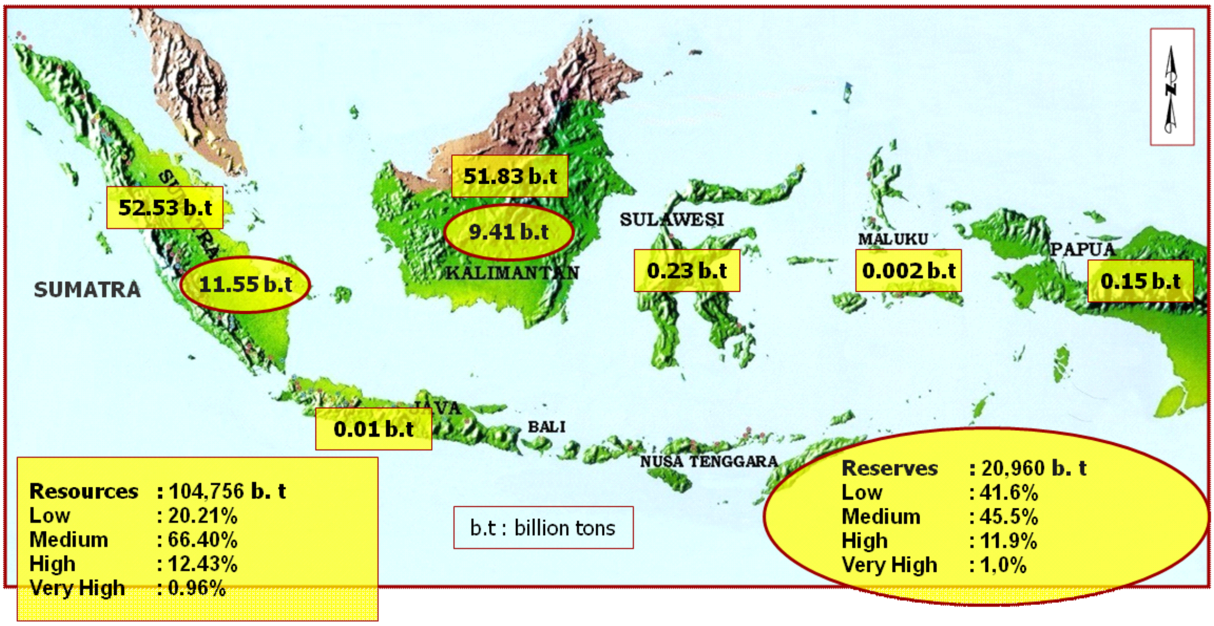
The distribution and quantity of coal resources and reserves based on the PP No 45/2003 is illustrated in Figure 1. Many of this coal reserves are deposited in areas of shallow overburden and can be recovered by strip mining at a modest cost. Although mining is relatively inexpensive, its high moisture content of LRC increases the cost of transportation and also creates unacceptable handling problems for both the coal producer and downstream consumers. Most of these low calorie coals

Received : 28 December 2010, first revision : 19 January 2011, second revision : 07 February 2011, accepted : February 2011

have not been utilized. Consequently, this coal should be consumed by electric generating stations located at or near the mine. The potential markets for low calorie coals can be enhanced if the coal firstly dewatered and upgraded prior to shipment to produce coal with heating value comparable to the high rank coal which has the limitation of reserves.

REVIEW OF COAL UPGRADING PROCESS

Lignite or sub-bituminous coals are commonly characterized as low rank coal (LRC). There is a growing demand of using these coals especially for power generation. In general, the direct use of LRC results in higher costs of reducing emissions or in lower efficiency and, consequently, higher



Source: Geological Agency, 2009 in Kamandanu, 2010

Figure 1. Indonesian coal resources and reserves

greenhouse gas emissions. It can therefore be stated that, because reserves for low calorie coal is quite plentiful, it is important to intensify efforts that will make the coal usable in an acceptable manner in terms of energy efficiency and environmental protection.

The benefit of coal upgrading includes increased value added to coal, stabilized coal quality feed for power generation and other industries, increased combustion efficiency and reduced CO_2 emission. Coal upgrading depends on many factors includes technology, market, economics, environment, policy and strategic issues being prime. At the present time, coal upgrading primarily mainly involves drying prior to direct use as fuel for combustion boilers in power plants to reduce the surface moisture of the coal.

To obtain the most suitable technology to be applied to Indonesian low calorie coal, research on coal dewatering by some coal upgrading processes in laboratory scale were carried out. The results were evaluated based on the degree of dewatering and the calorific value increasing as references for dewatering technology application.

Comprehensive understanding of all the forms of moisture in LRC and processes of moisture removal or release does not yet exist. Moisture in coal is incorporated as water absorbed on the surface, water condensed in the pores, water of hydration of inorganic components such as clay minerals, and water released via the thermal decomposition of organic oxygen functional groups. LRC contain more natural bed moisture than coals of higher rank, reflecting the dependence of pore volume on rank, which shows a progressive decrease from peat through the bituminous coals

(Anon, 1995).

The water absorbed on the surface (surface moisture) can be easily removed by mechanical and thermal drying techniques. In contrast, water condensed in the pores of coal (inherent moisture) removal needs some more techniques because the application of mechanical and simple thermal drying methods would be ineffective. Several upgrading processes through water removal (dewatering) of LRC include: thermal evaporative, mechanical, hydrothermal (i.e. under pressure thermal non-evaporative) as well as combinations of mechanical and thermal dewatering (Cough, 1990 and Allardice et al, 2003).

Evaporative processes such as flash drying, rotary drum drying, steam tube drying, fluidized bed drying, swirl tube drying, solar drying, etc have been widely applied in commercial stage (Couch, 1990). The first commercial plant was in Germany in the 1920s. The evaporative processes are largely adaptations of proven technology used with bituminous coals to remove surface water and improve heat content. When these processes are used to LRC, the dried product retain several properties which present problems to most users. In evaporative drying processes, there is not only latent heat involved but also the sensible heat of the solids is involved as well. High temperature, required to remove the capillary water from coal. Therefore, it is very energy consuming, requiring up to 20% of the chemical bonded energy of coal (Bergins, 2003). Furthermore, pure evaporatively LRC shows significant size degradation, easily re-adsorb moisture when exposed to either high humidity or rain and self ignition remains problematic (Sakaguchi et al, 2008). To prevent the readsorption of the moisture after upgrading process, an evaporative drying process followed by the addition of tar or residue to seal and plug the pore of coal was introduced and developed by Kobe Steel Ltd., Japan, namely upgraded brown coal (UBC) process. This process upgrades LRC to a high quality solid fuel with low moisture content and high calorific value (Deguchi et al, 1999).

Thereafter, mechanical processes have the potential advantage compared with evaporative drying and these processes seem to be more applicable for coals that have high moisture content and fibrous peat (Mursito et al, 2010). But, this method is complicated by the hydrophilic nature of LRC and the presence of water not just in the pores but also bonded chemically with the coal matrix, leading to its limited application. In practice such equipment would involve unacceptably high capital cost since the processes mostly operated at extremely high pressure, and consequently the processes are not currently in commercial contention. However, mechanical dewatering used in conjunction with other upgrading methods (e.g. MTE) has attracted attention to further reduce the water which proposed by Hulston et al (2004) and Artanto et al.(2009). Investigation work on both laboratory and pilot plant scales performed at temperature up to 280^{o}C and pressure up to 12 MPa. This process reduced moisture content to about 56 to 85%.

Among the drying processes, non-evaporative and under pressure treatments have the high competitive power due to the significantly lower energy requirement, where energy is not consumed by evaporation of liquid water, are favoured. Presently, dryings with simultaneous thermal upgrading are mostly used since these processes show the dramatically advantages. Non evaporative drying methods include drying with either steam or water. These processes not just remove the water but also alter the structure and chemical composition of the coal, thus truly upgrading the coal towards a more mature coal.

In this present study, three coal upgrading processes include upgraded brown coal (UBC) which is classified as thermal evaporative, hot water drying (HWD) and steam drying (SD) processes which are classified as thermal non-evaporative and under pressure were carried out using South Sumatera coal to study the characteristics of the raw and upgraded coals by those dewatering methods.

Upgraded Brown Coal (UBC)

The upgraded brown coal (UBC) process developed by Kobe Steel Ltd., Japan, is one of the evaporative drying processes. The UBC process upgrades LRC to high quality solid fuel on the basis of a slurry dewatering process. It has been developed and demonstrated as a pre-treatment for brown coal liquefaction (Shigehisa, et al., 2000). Indonesia under cooperation with Japan Center of Coal (JCOAL) has successfully developed UBC process on pilot scale with the capacity of 5 tons/day in Cirebon, West Java. The laboratory investigation of the process was started since 1993 and currently demonstration plant of 1,000 tons/day in Satui, South Kalimantan, Indonesia is underway.

UBC process is based on the oil-coal slurry dewatering process. The process conditions typically at temperature of 160°C and pressure of 0.35 Mpa, are much milder than those of other upgrading processes. This simplifies equipment required to carry out the process on an industrial scale reduces the cost of investment. Addition of low sulfur wax residue (LSWR) to the slurry at a concentration of about 1% is very important to prevent the re-absorption of moisture. The UBC can upgrade LRC into coal of 6,000 – 6,800 cal/g heating value, mostly through moisture content reduction technique. Moreover the UBC technique can be implemented as an intermediate coal process in order to produce a specific quality as required by the industries. The process is relatively simple and mild due to the non-chemical nature of the whole process. The final product is very stable and does not absorb atmospheric moisture.

The result of the feasibility study of the commercial based on pilot and demonstration scales held by Kobe Steel Ltd. proves that the UBC process will be able to produce a coal having a high calorific value and an upgraded coal which has a competitive price in the international market. The processing cost was calculated at a range of 8.2 - 10.3 US $/ton depends on moisture content of the coal of 25-50% (Tamura, 2010) as can be seen in Table 1.

as a liquid making this dewatering process less energy consuming by reducing the moisture content of LRC without evaporation of water. This is reached by heating the coal under pressure to reduce its moisture holding capacity, followed by separation of the water by mechanical means. Water removed as liquid may dissolve and leach out some water soluble inorganics with the wastewater, thereby reducing the amount of ash producing constituents in the upgraded solid. The temperature of above 150°C leads to decomposition of oxygen functional groups and results in formation of coals with higher carbon and lower oxygen contents. Removal of the hydrophilic oxygen functional groups permanently changes the surface characteristics to hydrophobic, thereby inhibiting water re-adsorption. Re-adsorption of water is also reduced through the collapse of the pore structure as well as blockage of pores by tar evolved during the treatment (Sakaguchi, 2008). The major advantage of the steam and hot water technologies is the quality and stability of the dried products, compared to the raw coal or to the products dried by other methods.

The HWD/SD process yields a product similar to higher rank coals. The dried coal has many of the beneficial characteristics of a bituminous coal. Moisture is low, the tendency to reabsorb moisture is reduced, and the coal is less prone to size

Table 1. UBC processing cost

Moisture, % ar	Feed t/day	Product t/day	Plant Investment* US $	Plant US $ Investment ** US $	Processing cost US $/t UBC
25	6,385	5,000	95	79	8.2
38	7,723	5,000	110	91	9.2
50	9,9592	5,000	121	101	10.3

Note:
* : including power generation
** : excluding power generation

Hot Water Drying (HWD) and Steam Drying (SD) Processes

Hydrothermal treatment, either with steam or water of LRC is a development from the Fleissner steam drying and the Evans-Siemon thermal dewatering process [Allardice, 2003]. During non-evaporative dewatering, the water in coal is removed

degradation, weathering, and spontaneous combustion (Chen, et al., 2000). Other benefits derived from HWD/SD include the reduction of sodium and to a lesser extent, reduction in sulfur content in the coal as moisture is removed (Cough, 1990). Sodium removal is important for many coals where high sodium is responsible for fouling in boilers.

EXPERIMENTAL

Coal Upgrading Processes

In this study the coal comes from Banko, South Sumatera was upgraded by UBC, HWD and SD processes. The UBC process was conducted in pilot scale with capacity of 5 tons/day in Palimanan, Cirebon, while the HWD and SD processes were conducted in laboratory scale.

- UBC Process
  The UBC pilot plant consists of five main sections. They were coal preparation, slurry dewatering, coal-oil separation, oil recovery and upgraded coal briquetting (Umar, et al., 2003). The outline of each section is as follows:
  - Coal preparation
    In this section, the raw coal was milled below 3 mm particle size by a hammer mill and stored in a coal bunker. The coal which was milled then was sent to a slurry maker tank.

  - Slurry dewatering
    Fine coal was mixed with kerosene and LSWR to prepare slurry in a slurry preparation tank. The mixing ratio of kerosene and coal was about 1.2 – 1.5 depending on the raw coal characteristics. The slurry was sent to a dewatering vessel via an evaporator where the moisture content of the raw coal was reduced by heating. More than 90% of the moisture in the coal was removed here. Afterwards, the dewatered slurry and the evaporated water were separated in a gas-liquid separator and the separated water was utilized as the heating of the evaporator after reaching its temperature level in a steam compressor by adiabatic compression. Dewatered slurry can be obtained by depressurizing down to atmospheric pressure in a slurry receiver. As dewatering proceeds in the evaporator, LSWR was preferentially adsorbed onto the inner surface of the pores disable the active points in the pores.

  - Coal-oil separation
    In this section, a screw decanter was applied to separate oil from dewatered slurry. Oil that occurred as free liquid in the slurry remained in the coal micro-pore. Separated cake was handled as a solid and sent to a dryer. A mechanical separator roughly separates free liquid in slurry.

  - Oil recovery
    Remaining oil in coal pores as the separated cake was recovered by steam tube dryer in this oil recovery section. Passing contact of the cake with a counter current flew of a carrier gas and indirectly heating of the steam tubes, evaporated the oil fraction producing dry coal. The evaporated oil was recovered by condensation and was reused as recycle oil for next slurry dewatering processes. Powdered UBC was discharged from the outlet of the dryer at about 170°C as the primary product.

  - Upgraded coal briquetting
    The hot UBC powder which was produced by steam tube dryer was sent to a double roll press briquetting machine to be briquetted without the addition of a binder. The UBC briquettes were stored in a product yard and some samples were taken to be characterized.

  Raw coal was ground to a grain size of under 3 mm and mixed with kerosene and LSWR to prepare slurry. The slurry was sent to a dewatering vessel where the moisture level was reduced by heating at the temperature of 150-160°C and pressure of 0.3-0.35 MPa (3-3.5 atm). The dewatered coal and oil were separated and were dried. The used oil was recovered and was reused as recycled oil for the next slurry dewatering. UBC product was in the form of briquette and was analyzed. The flow sheet of UBC process is illustrated in Figure 2.

- HWD Process
  The HWD process was conducted in laboratory scale at temperatures of 300ºC for one hour as the effective condition of process (Umar et al, 2007). The pulverized coal (ca. 1,500 g) was mixed with water (1:1) and was heated in an autoclave with 5,000 ml in capacity. The atmosphere in the autoclave was purge by N_2 at pressure of 0.1 MPa. The autoclave was heated at a rate of 3-4°C/min to reach the temperature of 300ºC and pressure of about 12 MPa. The upgraded coal was cooled, removed, dried and analyzed. Figure 3 shows the block diagram of HWD process.

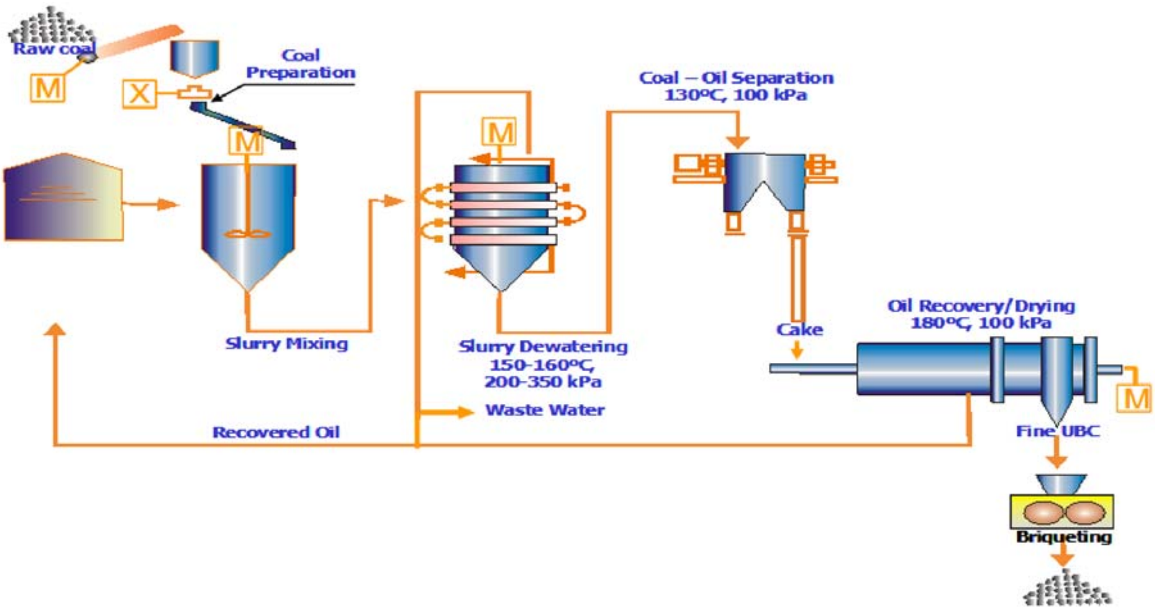


Figure 2. UBC process flow chart

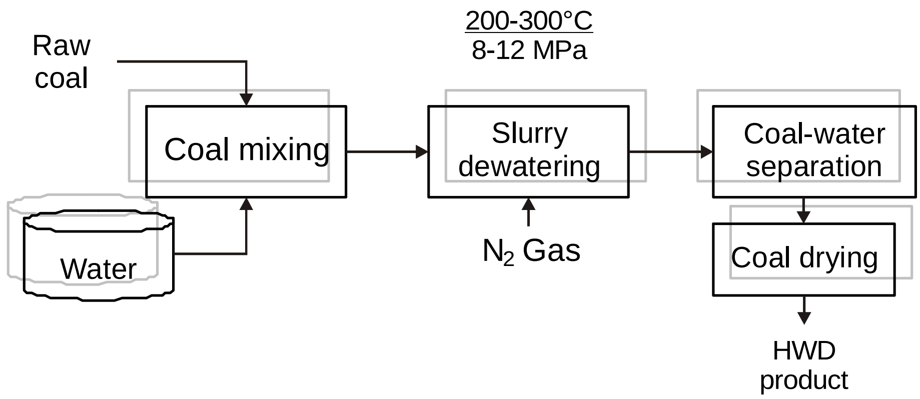


Figure 3. Block diagram of hot water drying process

- SD Process

  The SD process was conducted in laboratory scale. The different between HWD and SD processes is mainly depends on how coal feeding. SD process is affected by charging the autoclave with water and suspending the sample in a stainless steel basket above the water, at autoclave. While in HWD process, the autoclave is charged with slurry of coal and water.

  The process of SD was simple. The coal was heated under pressurized condition with saturated steam, and the water was removed in liquid form so that the latent heat of vaporization was not involved (Bongers, et al., 2003). Crushed and screened coal of -2+1 cm in size of about 1,500 g fed to an autoclave in 5,000 ml of capacity and was heated by steam at temperatures of 300ºC for one hour as the effective condition of process (Umar, 2004). As in the HWD process, the atmosphere in the autoclave was also purge by N_2 at pressure of 0.1 MPa. The autoclave was heated at a rate of 3-4°C/min to reach the temperature of 300ºC and pressure of about 12 MPa. The upgraded coal was cooled, removed and analyzed.

Characterization of the Raw and Upgraded Coals

The characterization of the raw and UBC upgraded coals was carried out based on chemical and combustion characteristics. Chemical analysis was based on proximate, ultimate, calorific value and functional froup analyses, whilst combustion properties based on differential thermal analysis (DTA) and thermo gravimetry (TG).

· Degree of dewatering
  The parameter of dewatering was evaluated by the moisture content data of the raw and upgraded coals. The degree of dewatering is defined by Eq. 1:

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

where: M was the moisture content (%) in the coals in air dried basis (adb).

- Proximate and ultimate analyses
  The proximate analysis covered the determination of moisture, ash and volatile matter and the calculation of fixed carbon. While the ultimate analyses is the determination of carbon and hydrogen in the material, as found in the gaseous products of its complete combustion, the determination of sulphur, nitrogen and ash in the material as a whole, and the calculation of oxygen by difference. These analyses were conducted according to the ASTM Standard Method (Annual Book of ASTM Standard, 2005).

- Calorific value
  The calorific value was determined by heating the coal sample in a bomb calorimeter at standard condition. The calorie produced, was calculated from the difference between initial and final combustion temperature with some correction. Calorific value was expressed in cal/g and is reported as gross calorific value in air dried basis.

- Functional groups
  A Fourier transform infrared spectrometer (HORIBA FT-720) in resolution of 1 cm^{-1} was used to study a change in functional group properties caused by the upgrading processes. The sample was measured by a KBr pellet method in spectrum from which 3000-2700 cm^{-1} and 1800-1500 cm^{-1} were assigned to the aliphatic hydrogen (C-H functional group) and oxygen containing structures (C=O functional group), respectively (Miura et al, 2002).

  The following parameters were defined as the ratio of deconvoluted peak area, $R_{CH3/CH2}$, $R_{ar/al}$, $R_{COOH/ar}$, and $R_{CO/ar}$, were defined as the ratios of methyl/methylene, aromatic/aliphatic, carboxyl/aromatic, and carbonyl/aromatic, respectively. Carbonyl groups include ester, carboxyl, and other carbonyl group such as ketone (Mahidin et al, 2002).

  $R_{CH3/CH2}$ = 2965 cm^{-1} band/2920 cm^{-1} band
  $R_{ar/al}$ = 1615 cm^{-1} band/(total of 2965, 2920, 2895, 2875, 2850 cm^{-1}, five bands)
  $R_{COOH/ar}$= 1710 cm^{-1} band/1615cm^{-1} band
  $R_{CO/ar}$ = (total of 1770, 1710, 1700, 1655 cm^{-1}, four bands)/1615 cm^{-1} band

- **Combustion characteristics**
  Understanding of the coal combustion characteristics aids in design and maintenance of the boiler. The good design helps to maximize combustion efficiency and assists in reducing carbon particle emissions. Thermal analysis data can be applied not only to the characterization of different coals, but also to the evaluation of combustion performance at high temperatures and heating rates. One of these techniques is differential thermal analysis (DTA) and thermo gravimetry (TG).

  DTA and TG, the monitoring respectively of the differentiation of heat release and the relative weight loss as a function of temperature, have been shown to be an effective tool to study combustion behavior of coal (Crelling, et al., 1992). Since only a small size of the sample is required to analyze, the combustion profile is most useful to evaluate the burning properties of fuel when only a small amount of samples is available or when it is impractical to test large quantities of fuel at existing installation (Ma et al, 1989).

  The DTA and TG analysis were carried out using a Shimadzu DTG-60 apparatus. The sample with weight of ca. 5 mg and particle size of less than 75 µm (passed through 200 mesh screens) was placed in a platinum cell at an airflow rate of 25 ml/min and heating rate of 10°C/min. The maximum experimental temperature was 800 °C (Usui et al, 2004). From the DTA-TG curves, a number of combustion parameters can be derived, such as ignition temperature (T_{ig}), maximum combustion rate temperature (T_{max}), maximum combustion rate (R_{max}), char burn out temperature (T_{bo}) and ash yield.

T_{max} was the temperature at which the maximum rate occurred and R_{max} from the DTA curve relates to the maximum combustion rate. The peak value of R_{max} of the DTA curve, give an indication of the intensity of combustion. Mass loss on the TG curve represents the amount of coal burned out. R_{max} is defined as in Eq. 2:

$R_{max}=[d(TGA)/dt]_{T=Tmax}$ .... (2)

Char burn out temperature (T_{bo}) was defined as the temperature at the minimum DTA peak after T_{max}, while ash yield a ratio of the sample weight remaining at the temperature of 800^oC (TG_{af}) to initial state (TG_{Ai}), as given in Eq. 3:

Ash Yield = TG_{Af}/TG_{Ai} x 100% .... (3)

RESULTS AND DISCUSSION

Change in Chemical Characteristics

The change in chemical characteristics due to coal upgrading processes is illustrated by the proximate, ultimate, calorific value and functional group analyses result of the coals. Table 2 shows the results of proximate, ultimate and calorific value of both the raw and upgraded coals.

From Table 2, it can be seen that inherent moisture of the raw coal (17.09%) decreased significantly after all of coal upgrading processes. The SD process reduced moisture content more than that of the UBC and HWD processes, consequently the degree of dewatering of the upgraded coal by the SD process was also the highest (Figure 4).

The highest degree of dewatering of the SD process (91.40%) than that of the UBC and HWD processes (88.99% and 88.12%, respectively), shows that the use of Banko coal in solid form was more effective than that in slurry form either with kerosene or water. Different from the previous study by using Berau coal from East Kalimantan with inherent moisture of 18.03% (Umar et al, 2007), the UBC process produced upgraded coal with the lowest degree of dewatering compared with that of the HWD and SD processes. It could be understood because the UBC process was

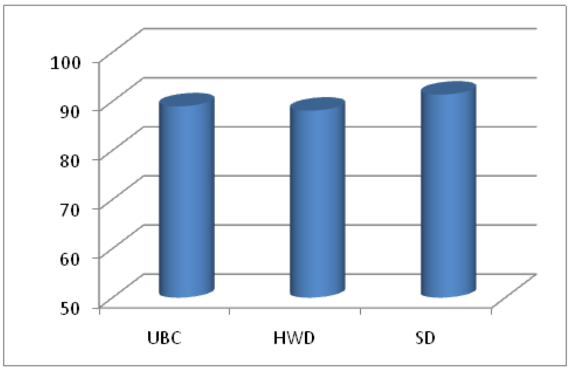


Figure 4. Coal upgrading degree of dewatering

Table 2. Analysis results of the raw and upgraded coals

	Raw	UBC	HWD	SD
Total Moisture, Wt% ar	25.32			
Moisture, Wt % adb	17.09	1.88	2.03	1.47
Ash, Wt % db	4.39	3.88	3.31	2.12
Volatile Matter, Wt % db	47.58	49.97	42.30	42.27
Fixed Carbon, Wt % db	48.03	46.15	54.39	55.61
Carbon, Wt % daf	70.99	72.89	79.09	77.79
Hydrogen, Wt % daf	6.28	6.15	6.30	6.40
Nitrogen, Wt % daf	0.88	0.89	0.75	0.76
Sulphur, Wt % daf	0.41	0.36	0.35	0.39
Oxygen, Wt % daf	21.44	19.71	13.51	14.66
Calorific value cal/g ar	4,901	6,919	6,929	7,085

Notes:
Adb : air dried basis Daf : dry ash free
Db : dry basis Ar : as received

conducted in lower temperature and pressure. contrary, if it is applied to the Banko coal, the degree of dewatering of UBC process higher than that of the HWD process (73.32 and 91.24%, respectively). The SD process indicates the highest degree of dewatering for both previous and present studies by using Berau and Banko coals, respectively.

As the reduction of moisture contents in the coal, the calorific value of the upgraded coals was also increased. Calorific value of the upgraded coals in as received (ar) basis is equal to the calorific value in air dried basis, if it was assumed that free moisture of the upgraded coals was zero. The calorific value of the upgraded coal by the SD process was also the highest than that of the UBC and HWD processes (Figure 5) in connection with the results of moisture contents. The calorific value of the upgraded coal by the UBC process was the lowest i.e. 6,919 cal/g ar. In this point of view, it can be concluded that all of the coal upgrading processes could be applied to the low calorie Banko coal to produce coal with high calorie (6,100-7,100 cal/g adb, according to the Indonesian Government Regulation No 45 year 2003).

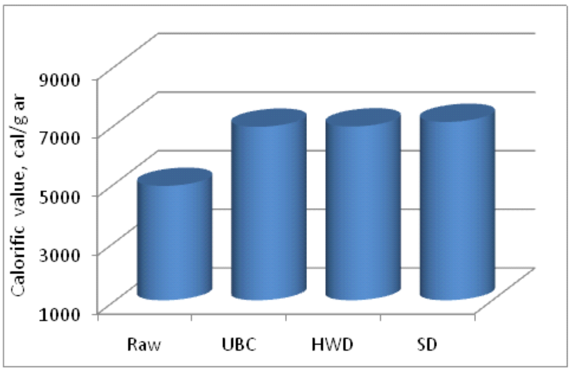


Figure 5. Calorific value of the raw and upgraded coals

The ash content of all upgraded coals significantly decreased compared to that of the raw coal. The highest reducing of ash content was reached by the SD process. As general explanation, the upgrading processes should not significantly affected by the ash content, due to the upgrading processes were focused on the reduction of moisture content. The increasing of ash content mostly was influenced by the impurities characteristics and the decrease of the coal mass. The UBC process produced coal with the highest ash content compare to that of HWD and SD processes. The high ash content of the UBC upgraded coal was caused by the LSWR pluged the coal pores and was remained as ash after combustion.

The volatile matter of upgraded coal by UBC process was increased. The HWD and SD processes, resulted in lower volatile matter. It was believed that the high temperature and pressure of devolatilization such as decarboxylation and dehydration was taken place (Mahidin et al, 2003).

The fixed carbon of the upgraded coals by HWD and SD processes was increased, while the fixed carbon was decreased by the UBC process . The decreasing of fixed carbon in the UBC upgraded coal caused by the significantly increased in the volatile matter. It might be explained that the LSWR which was added during UBC process plugged in the coal pore and it was detected as the volatile matter. The increasing of the fixed carbon for both upgraded coals by HWD and SD processes was due to the significantly decreased of moisture and slightly decreased of volatile matter contents.

The carbon content of the upgraded coals was increased for all of the UBC, HWD and SD processes. The slightly increased of the carbon content of the UBC upgraded coal was in connection with the decreasing in fixed carbon of the coal. The hydrogen content of the UBC upgraded coals was decreased, while by the HWD and SD processes it was slightly increased. The nitrogen content of the UBC upgraded coal was nearly equal to or slightly less than that of the raw coal. Whilst the upgraded coal by HWD and SD processes it was slighly decreased. So that the sulphur contents, all of upgraded coals show slightly decreased or equal to the raw coal. It could be concluded that all of the coal upgrading applied in this study hardly affect the hydrogen, nitrogen and sulfur contents.

The effect of upgrading process on the FTIR spectrum obtained for the raw and upgraded coals by UBC, HWD and SD processes is shown in Figure 6. The peaks of the upgraded coals were lower than that of the raw coal at the selected ranges of 3,000-2,750 cm^{-1} and 1,800-1,500 cm^{-1} which correspond to the structure of aliphatic hydrogen (C-H functional group) and the oxygen containing structures (C=O functional group) respectively. The peak of the UBC upgraded coal, higher than that of the HWD and SD upgraded coals. The HWD and SD processes which were carried out at higher temperature and pressure, several chemical constituents in the coal such as carboxyl, hydroxyl and ether functional groups, besides aliphatic and aromatic hydrocarbons are decomposed producing CO_2, H_2O, CO, CH_4, H_2, etc. and then were expelled from the coal. So that, the C-O and C=O of the coal were decreased. The parameters of the raw and upgraded coals in different upgrading processes are summarized in Table 3.

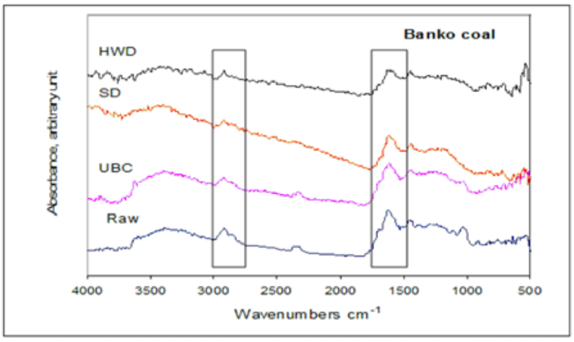


Figure 6. FTIR spectra for the raw and upgraded coals

Table 3. Parameters based on FTIR spectra of raw and upgraded coals

Coal	$R_{CH3/CH2}$	$R_{ar/al}$	$R_{COOH/ar}$	$R_{CO/ar}$
Raw	0.23	1.03	0.05	0.32
UBC	0.26	2.09	0.05	0.29
HWD	0.27	2.03	0.08	0.24
SD	0.28	3.95	0.06	0.26

As can be seen in Table 3, the $R_{CH3/CH2}$ values of all upgraded coals were increased and the methyl/methylene ratio generally increased when LRC was upgraded (Mahidin, 2002). The HWD and SD upgraded coals the R_{CH_3/CH_2} value was slighly higher than that of the UBC upgraded coal. This was caused by the lower temperature of the UBC process compared with those of the HWD and SD processes. The $R_{ar/al}$ values of all of the upgraded coals were significantly higher than that of the raw coal. The SD upgraded coal shows the highest $R_{ar/al}$ value (3.95) compare to that of raw, UBC and HWD upgraded coals (1.03; 2.09 and 2.03, respectively). The high the $R_{ar/al}$ value, the high the aromaticity of the coal. It indicates that the SD upgraded coal is similar to the high rank coal which has higher aromaticity than alifaticity (Ohki et al, 1999).

The values of $R_{COOH/ar}$ almost did not change during the upgrading processes. By the UBC process, the upgraded coal it was exactly the same and by the HWD and SD processes it was slightly increased. The difference (slightly increased and exactly the same) might be concluded that those upgrading processes were not affected the value of the $R_{COOH/ar}$.

The $R_{CO/ar}$ for upgraded coals decreased from 0.32 become 0.29; 0.24 and 0.26 after coal upgrading by UBC, HWD and SD processes, respectively. The lowest decrease of the $R_{CO/ar}$ value, was reached by UBC process. It was due to the low temperature process that applied in the process, so that the released of the carbonyl group in the coal was not completed yet.

Change in Combustion Characteristics

DTA curves of the raw and upgraded coals are shown in Figure 7. The curves illustrated heat differentiation which was released during the test. There were three DTA peaks for all of the raw coals. The first and second DTA peaks appeared around 60°C and 330°C due to the vaporization of moisture and combustion of volatile matter. The third peak represented the combustion of char.

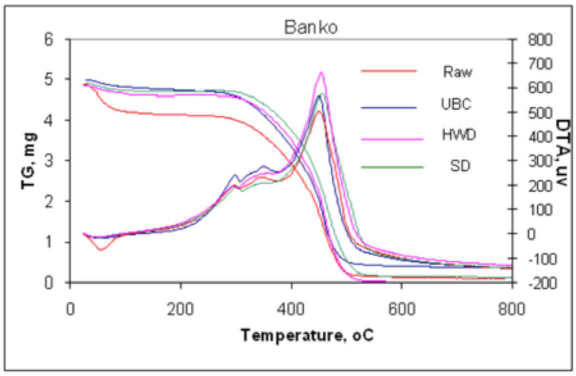


Figure 7. DTA-TG for the raw and upgraded coals

For all of the upgraded coals, the first peaks significantly decreased because of moisture removal during upgrading processes. The second peaks, corresponded to volatile matter, removal didnot change during the UBC process because this process was conducted at mild temperature but slightly decreased by the HWD and SD processes.

Furthermore, the third peaks that show the highest, all of the upgraded coals were significantly increased. The increase of third DTA peaks of all of the upgraded coals characterized that the major heat released in combustion of upgraded coals was taken place. Based on this fact, it seems that the heating values of the upgraded coals were higher than that of raw coals. Generally speaking, the HWD result upgraded coal with the highest DTA maximum peak than that of the upgraded coal by the SD and UBC processes.

The TG curves of raw and upgraded coals illustrated the relative weight loss. Weight loss below 150°C was related to moisture removal , and that above of 150°C was due to the combustion of volatile matter and char. Table 4 shows the summary obtained for combustion parameters from the DTA-TG. A bituminous coal from Kaltim Prima Coal (KPC) was also described as reference (Mahidin et al, 2002).

Table 4. Combustion parameters based on DTA-TG analysis of the raw and upgraded coals

Coal	T_{ig} °C	T_{max} °C	T_{bo} °C	Rmax mg/min
Raw	282	453	789	0.30
UBC	289	449	783	0.34
HWD	308	453	783	0.39
SD	312	456	785	0.35
KPC	327	495	583	0.28

The T_{ig} value corresponds to the ignition temperature of the volatile matter. In this study, the T_{ig} of all of the upgraded coals increased. In UBC upgraded coal, the T_{ig} was equal to or slightly increased. While the HWD and SD upgraded coal significantly increased. The increasing of T_{ig} in the HWD and SD upgraded coals was discribed to the decreasing of the volatile matter since the ignition for HRC was controlled by the volatile content. However, it was difficult to cite the general comparison between HRC and LRC because the ignition of LRC was largely influenced by the reactivity of the oxygen. In general, those coals with low ignition temperature and high mass loss in the low temperature range can be considered as easy to ignite and burn out.

T_{max} relates to coal reactivity. Reactive coals have a lower of T_{max}. , it can be seen From Table 4 that the T_{max} of upgraded coals due to the upgrading process was almost no change (UBC upgraded coal was slightly decreased, the HWD upgraded coal was exactly equal and SD upgraded coal was slightly increased).

R_{max} indicates the maximum combustion rate. All of upgraded coals have the R_{max} was higher than that of the raw and KPC coals. The highest R_{max} was reached by upgraded coal through the HWD process, followed by upgraded coals by SD and UBC processes in relevant to the DTA maximum peak. It expressed that the upgraded coals were easy to be burnt due to high calorific value and low moisture content (Ohki et al, 1999).

The T_{bo} reflects that the char characteristic was almost no change or slightly decreased due to upgrading treatment. Compared with the combustion characteristics of KPC coal that characterized as high calorie coal, the char characteristic of upgraded coals was widely different from the char characteristics of high rank natural coal which was shown by higher T_{max} and T_{bo}.

Since the area under the DTA curve was interpreted as the heat change during the whole process, the DTA curve can be used to estimate heat which was released during combustion process and corresponds to the high heating value of coal. In this study, the relationship of the high heating value calculated (HHVcalc, MJ/kg) and area under DTA curve, *A* (µV. sec) is represented as the equation 4 with correlation coefficient (R) of 0.775 and the results can be seen in Table 5.

Table 5. Calculated and measured HHV of the raw and upgraded coals

	DTA max peak, µv	Time, sec	TGAi, mg	A/TGAi, µv sec/mg	HHV measured MJ/kg	HHV calc. MJ/kg
Raw	**504**	**2530**	**4.88**	**261,295.1**	**21.13**	**23.28**
UBC	567	2486	5.00	281,912.4	26.42	24.69
HWD	662	2519	4.86	343,123.0	27.76	28.87
SD	577	2530	4.91	297,313.6	27.29	25.75

HHV calc = 5.43896 + ($6.83011.10^{-5}$ x A/TGAi) ... (4)

Table 5 indicates that the HHVs which were calculated from DTA analyses different from the HHVs which were measured using bomb calorimeter according to ASTM Standard D-3286. It might be explained by the deviation of equations 4 which was used. It was reflected by low correlation coefficient (0.775) between the DTA max peak, time, TGAi and HHV measured. It can be seen that the HHVs of the upgraded coals by the HWD and SD processes were higher rather than that of the raw and upgraded coals by the UBC process, based on higher peaks of the upgraded coals.

CONCLUSIONS

Various coal upgrading of UBC, HWD and SD processes which were applied to a low calorie Banko coal indicate that:

– the inherent moisture of the upgraded coals was significantly decreased. Consequently, the specific energy of all upgraded coals also significantly increased. The SD process reduced moisture content more than that of the UBC and HWD processes;

– the FTIR spectrum obtained showed that the peaks of the upgraded coals were lower than that of the raw coal at the selected ranges of 3000-2750 cm^{-1} and 1800-1500 cm^{-1} and correspond to the structure of aliphatic hydrogen (C-H functional group) and the oxygen containing structures (C=O functional group) respectively;

– the T_{ig} value which corresponded to the ignition temperature showed that all of the upgraded coals increased. It reflected that the coals have lower tendency to spontaneous combustion compare to that of the raw coal;

– the upgraded coals had better combustion characteristics it was shown by the R_{max} values that indicates the maximum combustion rate. All of the upgraded coals was higher than that of the raw coals;

– technically, the LRC upgrading by the HWD and SD processes produced upgraded coal with better characteristics than that of the upgraded coal by the UBC process. The inherent moisture contents were lower and the calorific values of the upgraded coals were higher and have better combustion characteristics. The SD process produced upgraded coal with the highest calorific value than that of the UBC and HWD processes in relevant to the moisture content of the upgraded coals.

REFERENCES

Allardice, D. J., Clemow, L. M., Favas, G., Jackson, W. R., Marshall, M. and Sakurovs, R., 2003. The characterization of different forms of water in low rank coals and some hydrothermally dried products, *Fuel, 82*, p. 661 – 667.

Annual Book of ASTM Standards, 2005. Copyright ASTM International, 05.06, Section Five; *Gaseous Fuels, Coal and Coke*, 100 Barr Harbor Drive, West Conshohocken, USA.

Artanto, Y., McDonnell, E., Verheyen, T. V., Adeloju, S. and Chaffee, A. L., 2009. The remediation of MTE water by combined anaerobic digestion and chemical treatment, *Fuel, 88*, p. 1786–1792.

Bergins, C., 2003. Kinetics and Mechanism During Mechanical/Thermal Dewatering of Lignite, *Fuel, 82*, p. 355–364.

Bongers G. D., Jackson, W. R. and Woskoboenko, F., 2003. Pressurised Steam Drying of Australian Low-Rank Coals Part 2. Shrinkage and Physical Properties of Steam Dried Coals, Preparation of Dried Coals With Very High Porosity, *Fuel Processing Technology 64-2000*, p. 13–23.

Chen, Z., Wu, W. and Agarwal, P. K., 2000. Steam-Drying of Coal. Part 1. Modeling The Behavior of a Single Particle, *Fuel, 79*, p. 961–973.

Couch, G. H., 1990. *Lignite Upgrading*, IEA Coal Research. IEACR/25.

Crelling, J. C., Hippo, E. J. and Woerner, B. A., 1992. Combustion Characteristics of Selected Whole Coals and Macerals, *Fuel, 71*, p. 151-158.

Deguchi, T., Shigehisa, T. and Shimasaki, K., 1999. Study on Upgraded Brown Coal Process for Indonesian Low Rank Coals, *Proc. International Conference on Clean and Efficient Coal Technology in Power Generation, Indonesia*, p. 176-180.

Hulston, J., Chaffee, A. L., Bergins, C., and Strauâ, K., 2004. Comparison of Physico-Chemical Properties of Various Lignites Treated by Mechanical Thermal Expression, *Coal Preparation, 25-4*, p. 269-294 .

Indonesian Government Regulation Number 45 Year 2003 about the Rates of Non Tax Revenues at the Department of Energy And Mineral Resources.

Kamandanu, B., 2010. Potential for Government Incentives in Developing Indonesian Coal Resources, *the 2nd Coaltrans Upgrading Coal Forum, The Grand Hyatt Jakarta, 21-22 September 2010.*

Lignite of America, 1995. Chapter 10, Beneficiation of Lignite, *Coal Science and Technology*, 23, p. 451-504.

Ma, S., Hill, J. O. and Heng, S. J., 1989. Study of the Combustion Characteristics of Victorian Brown Coal, *Journal of Thermal Analysis, 35*, p. 1985-1996.

Mahidin, T., Usui, H., Ishikawa, S, and Hamdani, 2002. The Evaluation of Spontaneous Combustion Characteristics and Properties of Raw and Upgraded Indonesian Low Rank Coals, *Coal Preparation, 22*, p. 81-91.

Mahidin, T., Ogaki, Y., Usui, H. and Okuma, O., 2003. The Advantages of Vacuum-Treatment In the Thermal Upgrading of Low-Rank Coals on the Improvement of Dewatering and Devolatilization, *Fuel Processing Technology, 84*, p. 147– 160.

Miura, K., Mae, K., Ashida, R., Tamura, T. and Ihara, T., 2002. Dewatering of Coal Through Solvent Extraction, *Fuel, 81*, p. 1417-1422.

Mursito, A. T., Hirajima, T. and Sasaki, K., 2010. Upgrading and Dewatering of Raw Tropical Peat by Hydrothermal Treatment, *Fuel, 89*, p. 635–641.

Ohki, A., Xie, X. F, Nakajima, T., Itahara, T. and Maeda, S., 1999. Change in Properties and Combustion Characteristics of an Indonesian Low Rank Coal Due to Hydrothermal Treatment, *Coal Preparation, 21*, p 23-34.

Sakaguchi, M., Laursen, K., Nakagawa, H. and Miura, K., 2008. Hydrothermal Upgrading of Loy Yang Brown coal — Effect of Upgrading Conditions on The Characteristics of The Products, *Fuel Processing Technology, 89*, p. 391-396.

Shigehisa, T., Deguchi, T., Shimasaki, K. and Makino, E., 2000. Development of UBC Process. *Proc. International Conference on Fluid and Thermal Energy Conversion*, Indonesia.

Suhala, S., 2010. Current and Future Coal Situation : Supply & Demand and Price Mechanism, Presented at : *Focus Group Discussion on Upgraded Brown Coal*, Kartika Chandra, Jakarta.

Tamura, M., 2010. New Utilization Technologies for Low Rank Coals, *Proc. 1st International Symposium on Low Rank Coals, Innovation and Technologies for a Sustainable Future*, Grand Hyatt Melbourne, Australia.

Umar, D. F., Suganal, Priyono, H., Nuroniah, N., Sodikin, I., Supriatna, W., Rijwan, I., Rustomo, G., Syahrial, Hernawati, T., Astiti, M. W., Atmini, S., Paidi, Rohayati, T., Agustiana, L., Somadi, E. dan Aat, 2003. Uji Coba Peningkatan Kualitas Batubara Peringkat Rendah Dengan Proses UBC (Upgraded Brown Coal) Palimanan, Cirebon, *Laporan Litbang tekMIRA No: 09*.

Umar, D. F., 2007. Development of Low Rank Coal Upgrading and Upgraded Brown Coal Water Mixture Preparation Technologies, Kobe University, *Doctoral Dissertation*.

Usui, H., Mahidin, T. and Umar, D. F., 2004. Upgrading by Kobe University Process and Control of Spontaneous Combustion of Low Rank Coals, *Proc. 21st International Pittsburgh Coal Conference, Osaka*, Japan.