UPGRADING OF INDONESIAN LOW RANK COAL BY STEAM DRYING METHOD

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ABSTRACT

Experiments were carried out to produce upgraded coals with low moisture content by steam drying method. An Indonesian low rank coal, Berau coal with moisture content of 16.13% and calorific value of 5324 cal/g in air dried basis (adb) was treated by steam drying at temperature of 225 - 300°C in an autoclave to study process temperature effect. The results of proximate and ultimate analyses show that moisture content decrease by increasing the temperature process. The moisture content reduces to 0.86% and the calorific value increases up to 6760 cal/g in adb when the coal has been treated at the temperature of 300°C. Furthermore, the specific surface area and combustion characteristics of coals were influenced by the upgrading process.

1. INTRODUCTION

Since the energy crisis many years ago, many countries have attempted to look for alternative energies such as coal, nuclear, solar, wind, etc. These are being investigated and developed to reduce the dependencies on oil and gas. By considering the many advantages of coal and the weakness of other energies, coal is considered to be the first alternative to replace oil and gas in Indonesia.

A large amount of coal resources has found almost all over Indonesia and most of them are lignite and sub-bituminous coal which referred as low rank coal. Many of these low rank coal resources are deposited in areas of shallow overburden and can be recovered by strip mining at a relatively low. Although mining is relatively inexpensive, the fuel has a comparatively low heating value because of its high moisture content. Therefore, access to markets is limited by high transportation costs. Consequently, most low rank coals are consumed by electric generating stations located at or near the mine. The potential markets for low rank coal can be enhanced if the coal firstly dewatered and upgraded prior to shipment.

Many studies have been conducted on the upgrading of low rank coals. Willson et al (1987), have put forward the hot water drying (HWD) method in comparison with alternate methods (drying by a rotary kiln, superheated nitrogen, superheated steam and saturated steam). The high pressure condition of this upgrading process is a significant disadvantage. Usui et.al (1998), proposed the upgrading method by a combined process of vacuum drying and tar coating at low pressure. The additional of tar was needed to seal and plug the pore of coal to prevent the re-absorb of moisture after the upgrading process.

In this study, the upgrading technique by steam drying originally patented by Fleissner (1927), and developed by Koppelman, (1977) is investigated (Chari and Huettenhein,1985). This process removes significant amounts of moisture from low rank coal using heat and pressure. In simple terms, the low rank coal was cooked and aged so that the product becomes similar to bituminous coal. The major advantage of this technology is the quality and stability of the upgraded coal compared to the raw coal or to the upgraded coal by other methods [Chari and Huettenhein,1985]. The upgraded low rank coal has many of the beneficial characteristics of a bituminous coal. The moisture is low, the tendency to reabsorb moisture is reduced, and the coal is less prone to size degradation, weathering and spontaneous combustion.

Steam drying process heats coal to the temperature above 200°C for decomposing some of the carboxyl groups in the coal and releasing carbon dioxide. The carbon dioxide in turn expels moisture seals into the pores of the coal structure. This moisture is released in the form of liquid water. Water is also forced from the coal pores by differences in thermal expansion of water and coal. Because the removal of moisture is achieved without evaporating the water, the process has a potential to improve energy efficiency compared to evaporative process [Stanmore et al., 1982]. More importantly, the tars released during the process could coat the surface of the coal particles and prevent the re-absorb of moisture to the upgraded coal.

The steam drying process has been conducted using low rank coal from Berau, East Kalimantan. This coal has a high moisture content. Consequently, this coal has a low calorific value although it has a low ash content. This paper reports the effect of processing temperature towards the chemical and physical characteristics, including the combustion properties of the upgraded coals.

2. EXPERIMENT

The process of steam drying was conducted using an autoclave in 5 l/batch of capacity. The coal was heated under pressurized condition with steam, and the water is removed in liquid form so that the latent heat of vaporization is not involved [Klein, 1930]. About 200 grams of crushed and screened coal in the size between 1 to 2 cm and the weight of was fed to an autoclave. The coal was heated by steam under pressure up to 5.5 MPa an temperature range of 225 to 300°C to obtain the condition of steam drying process based on the moisture content reduction. After the autoclave was cooled down, the upgraded coal was removed, dried and analyzed (proximate analyses, ultimate analyses and gross calorific value according to the ASTM Standard).

The specific surface area (m^2/g) for both the raw and upgraded coals was tested by BET (*Brunauer-Emmet-Teller*) method using N_2 at 25°C. It was measured using SHIBATA APP-SA-100 based on the amount of nitrogen absorbed on the surface of the solid particle. The nitrogen flow rate was 150 ml/min. Before subjected to the test, coal samples were dried in the nitrogen atmosphere at 180°C.

To study the change in combustion properties, the differential thermal analysis-thermo gravimetric (DTA-TG) for both the raw and upgraded coals were tested using a Shimadzu DTG-60 apparatus. About 5 mg of sample with the particle size less than 75 µm (under 200 mesh screens) was placed in a platinum cell, heated at a rate of 10°C/min, with an air flow rate of 25 ml/min. The maximum temperature of combustion was 800°C. Combustion parameters, such as the ignition temperature (*T ig*), maximum combustion rate temperature (*T max*) and the maximum combustion rate (*R max*) were derived from the DTA-TG curves [Ohki et al., 1999].

3. RESULTS AND DISCUSSION

3.1 Change in Chemical and Physical Properties

3.1.1 Proximate and ultimate analyses

All steam drying treated run upgraded coals that were at more than one processing temperature exhibited linier decreases in moisture content with the increasing temperature. According to Murray et al [Murray and Evans, 1972], low rank coals contain appreciable amounts of oxygen, in the form of carboxylic acids and salts of carboxylic acids. This fact contributes to the low heating value due to their affinity to moisture. A carboxylic acid molecule is polar and can form hydrogen bonds with other polar molecules, in particular with water. The acidic oxygen contained in functional groups destroys at about 150°C. The formed carbon dioxide that was is considered to expell water from the coal pores in coal dewatering process.

The steam drying process at temperature of 300°C produces an upgraded coal with least moisture content as can be seen in Table 1. In this condition, the moisture content of the upgraded coal decreases significantly from 16.13% to 0.86% (adb). As the processing temperature increased of above 225°C, the extent of coal dehydration becomes increasingly prominent which is illustrated by the decreasing of moisture content from the coal. It is evident that the temperature increasing can greatly reduce the moisture content. The re-

Table 1. Proximate and ultimate analysis of the raw and steam drying upgraded coals

Coal	Moist. Wt% (adb)	Ash Wt% (db)	VM Wt% (db)	FC Wt% (db)	C Wt% (daf)	H Wt% (daf)	N Wt% (daf)	S Wt% (daf)	O Wt% (daf)	Calorific value cal/g (adb)
Raw	16.13	7.58	44.35	48.07	69.68	6.46	1.84	0.59	21.43	5324
SD, 225ºC	1.39	4.59	44.06	51.35	70.90	5.23	1.51	0.39	21.97	6315
SD, 250ºC	1.24	4.43	43.77	51.80	71.36	5.26	1.50	0.24	21.64	6441
SD, 275ºC	1.09	3.98	42.47	53.55	73.25	4.97	1.52	0.34	19.93	6501
SD, 300ºC	0.86	3.72	41.05	55.23	73.56	4.78	1.53	0.30	19.83	6760

Note: adb: air dried basis; db: dry basis; daf: dry ash free; SD: steam drying

ducing of moisture content causes the calorific value of the coal increases from 5324 cal/g in raw coal to 6760 cal/g in upgraded coal when the coal was steam drying treated at the temperature of 300°C. Due to the increase in temperature, the compressive yield stress on the coal and, hence, the resistance of the coal structure to deformation is decreased, and the coal network becomes softer [Guo et al., 1998].

When the upgrading temperature of the steam drying process is increased, the ash content and the volatile matter (VM) of the coal were significantly decreased. It could be explained; that the heat may dissolve a part of impurities in the coal due to the decreasing of ash content. The decomposition product also enhances the devolatilization process that reduces gases and vapor which interpreted as the volatile matter.

The nitrogen and sulfur content of the upgraded coals are slightly reduced compared with the raw coal. The sulfur content decreases from 0.59% in raw coal to 0.30% on a dry ash free basis by steam drying process at 300°C. According to Couch [1990] as well as coal dewatering, the other benefit derived from the steam drying process is the reduction of the sulfur content.

3.1.2 Specific Surface Area

Coal upgrading process expects the specific surface area will be lower than that of the raw coal. Due to the shrinkage and plugging of coal pores by tars. The generated tars during steam drying process exuded to the coal surface and sealed the micropores on the surface to reduce the specific surface area. Therefore, the moisture that was released during heating can be removed permanently Table 2 shaves that. The specific surface area for all of upgraded coals is lower than that of the raw coal. The coal particle size is hardly effect the specific surface area.

Table 2. The BET surface area of raw and steam drying upgraded coal

Coals	Specific surface area, m^2/g		
	-200 mesh	-60+200 mesh	-60 mesh
Raw	6.65	5.78	5.12
Steam drying, 225ºC	3.88	3.72	3.27
Steam drying, 250ºC	2.30	2.17	2.16
Steam drying, 275ºC	1.66	1.64	1.16
Steam drying, 300ºC	1.03	0.92	0.78

3.2 Combustion Characteristics

DTA and TG curves of the raw and 300ºC steam drying treated coals are given in Figures 1 and 2 respectively. The curves indicate the differentiation of released heat and relative weight loss during testing time. Combustion of LRC usually corresponds to two or more DTA peaks due to the complicated structure of the coal [Ma et al., 1989]. In the case of the raw coal, there are three DTA peaks, as can be seen in Figure 1.

The first and the second DTA peaks appear at about 60°C (endothermic) and 330°C (exothermic) and are associated with the vaporization of moisture and combustion of volatile matter respectively. In case of the coal was upgraded at the temperature of 300°C, the first peak (which is a trough due

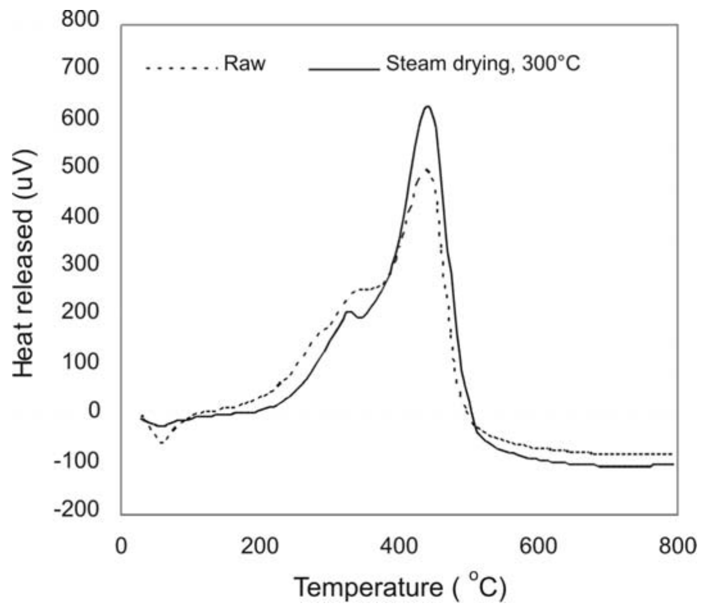

Figure 1. DTA curve for the raw and upgraded coals

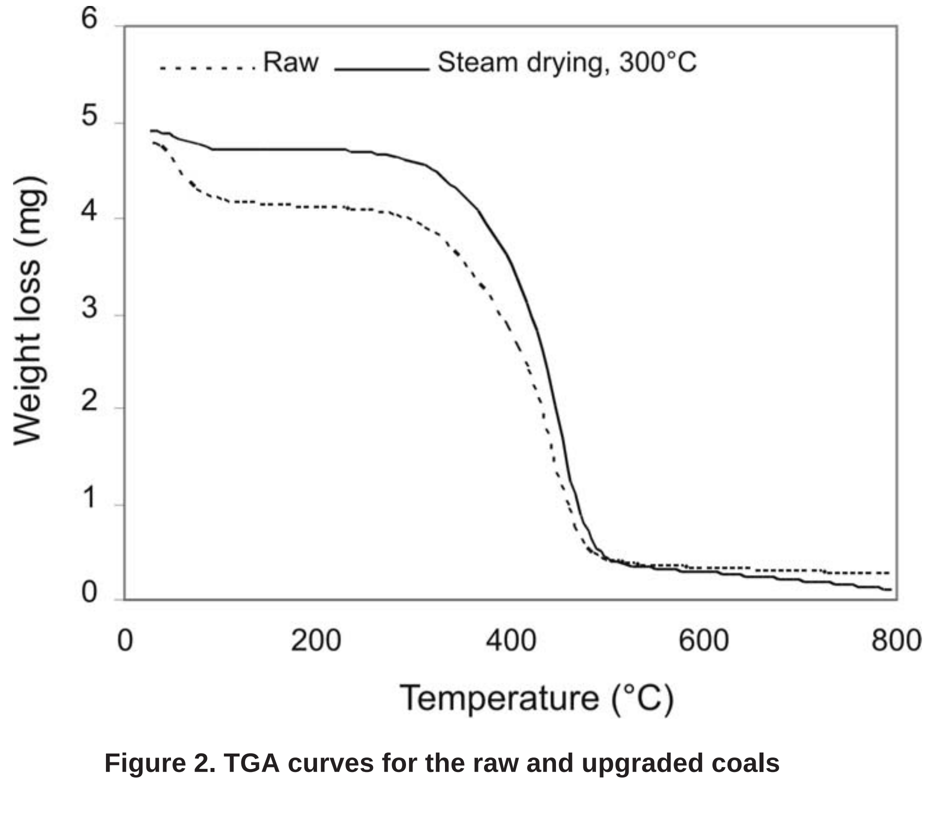

Figure 2. TGA curves for the raw and upgraded coals

to being endothermic) decreases since the moisture content has decreased. The second peak, which corresponds to removal of volatile matter [Mahidin et al., 2003], slightly decreases after steam drying process due to the reduction in volatile matter. The third DTA peak (max = 505 µV), which occurs at about 437°C (exothermic) in case of the raw coal, represents the combustion of char. This peak in the coal upgraded at 300ºC was increased significantly (max = 637 µV) and occurred at the temperature of 443°C. The increase of the third DTA peak in the upgraded coal shows that a greater heat was releases during combustion. Therefore the calorific value of the upgraded coal is higher than that of the raw coal.

Several parameters that can be derived from DTA-TG analysis are summarized in Table 3 [Ohki et al., 1999]. The Kaltim Prima Coal (KPC), which referred as bituminous coal was also described as reference [Mahidin et al., 2003]. The *T ig* value, which corresponds to the ignition point of the volatile matter, was increased by increasing the steam drying temperature. In the case of the upgraded coal at the temp of 300°C, the *T ig,* which is initially 215°C (raw coal), increases to 285°C.

Table 3. Combustion parameters based on DTG-TA analysis of the raw and steam drying upgraded coals

Coal	*T ig* (ºC)	*T max* (ºC)	*R max* (mg/min)
Raw	215	437	0.29
Steam drying, 225ºC	263	436	0.29
Steam drying, 250ºC	273	435	0.33
Steam drying, 275ºC	278	448	0.32
Steam drying, 300ºC	285	443	0.35
KPC	327	495	0.28

The increase of T_{ig} is ascribed to the decrease of the volatile matter, since the ignition for higher rank coals are controlled by the volatile content. But it is difficult to quote general comparison of high and low rank coals, because ignition of low rank coal is influenced by the reactivity of the oxygen. Those coals with the low ignition temperature and high mass loss in the low temperature range are generally easy to be ignited and to be burntout.

T_{max}, whic relates to the the coal reactivity, T_{max} of reactive coal has a low value. It is recognized from Table 3 that T_{max} of upgraded coals due to the steam drying process change a little only smal or almost relatively the same. When it is compared with the combustion characteristics of the high rank coal (KPC), the char characteristics of the upgraded coal is very different from the char characteristics of natural high rank coal as shown in Table 3. KPC coal indicates high T_{max} is 495°C. Different from the case of T_{max} *and* T_{bo} the R_{max} value of the upgraded coal, which indicates the maximum combustion rate, is much higher than those of raw and bituminous coals. It can be said that it is easier for the upgraded coal to be burnt due to its of low moisture content and high calorific value.

4. CONCLUSIONS

Steam drying process has been studied for the Indonesian Berau low rank coal to obtain the processing temperature condition based on the decrease in moisture content. The processing temperatures investigated ranged from 225ºC to 300ºC.

The least moisture content was achieved at the processing temperature of 300°C. In this condition, the moisture content significantly decreased from 16.13% adb in raw coal to 0.86% in upgraded coal. Subsequently, the calorific value increases from 5324 cal/g to 6760 cal/g in adb.

The specific surface area of upgraded coals is lower than that of the raw coal. The shrinkage and plugging of coal pores caused by the tars that were generated during steam drying process have coat the upgraded coal surface effectively.

The steam drying process produces coal with better combustion characteristics than that of the raw coal. The *T ig* for all of the upgraded coals are higher than that of the raw coal, which can be considered as having lowered the susceptibility to self ignition. At the same time, *T max,* which reflects coal reactivity, is not significantly changed due to the steam drying process.

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