INDONESIAN MINING JOURNAL Vol. 18, No. 1, February 2015 : 11 - 17

COAL DE-ASHING BY SOLVENT EXTRACTION

PENURUNAN KADAR ABU BATUBARA DENGAN CARA EKSTRAKSI PELARUT

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ABSTRACT

Coal contains mineral matter that will be left as ash after coal is burned. Coal will be referred to as dirty coal if the ash content of the coal is high. High ash content is not preferred by consumers of coal users especially coal fired power plants, because ash content will produce fly ash and bottom ash that cause environmental problem. The process of ash content reduction by solvent extraction would produce coal with very low ash content (near zero) known as ash free coal (AFC). The study of ash content reduction was conducted by using Peranap coals that were taken from stockpile and mine site. The coals were then washed and separated into coals with low and high ash contents. The high ash content of coals from stockpile (46.02%) and mine site (25.02%) were then extracted using solvent. Three kinds of solvent have been tested, namely 1-methyl naphthalene, 1-1-1-methoxy ethoxy acetic acid and N-methyl 2 pyrolidynon. The results indicate that the ash content of coal derived from the stockpile decreased to 0.06% and coal from the mine site decreased to 0.11% by using 1-methyl naphthalene solution with a ratio of coal and solvent of 1: 6 (weight/weight).

Keywords: mineral matter, ash, solvent, extraction, ash free coal (AFC)

SARI

Semua batubara mengandung mineral matter. Residu dan mineral ini akan menjadi abu kalau batubara dibakar. Batubara akan disebut sebagai batubara kotor apabila kandungan abu dalam batubara tersebut cukup tinggi. Kadar abu yang tinggi tidak disukai konsumen pengguna batubara terutama pembangkit listrik tenaga uap karena kadar abu akan menghasilkan abu terbang dan abu bawah yang menyebabkan masalah lingkungan. Oleh karena itu perlu dilakukan penelitian penurunan kadar abu. Penelitian dilakukan dengan cara ekstraksi untuk menghasilkan batubara dengan kadar abu yang sangat rendah hampir mendekati nol yang disebut dengan batubara bebas abu. Penelitian ini menggunakan batubara Peranap yang diambil dari tumpukan dan tambang. Batubara tersebut kemudian dicuci dan dipisahkan antara batubara yang berkadar abu rendah dan batubara yang berkadar abu tinggi. Batubara dengan kadar abu tinggi yang berasal dari tumpukan dan tambang yang masing-masing mempunyai kadar abu sebesar 46,02% dan 25,43%, kemudian diekstraksi. Percobaan dilakukan dengan menggunakan 3 jenis pelarut, yaitu 1-metil naftalen, 1-1-1-metoksi etoksi asetik acid dan N-metil 2 pirolidinon. Hasil penelitian menunjukkan bahwa kadar abu batubara yang berasal dari tumpukan turun menjadi 0,06% dan batubara yang berasal dari tambang turun menjadi 0,11% dengan menggunakan larutan 1-metil naftalen dan perbandingan antara batubara dan pelarut 1:6 berat/berat.

Kata kunci: bahan mineral, abu, pelarut, ekstraksi, batubara bebas abu (BBA)

Received : 11 November 2014, first revision : 07 January 2015, second revision : 05 February 2015, accepted : February 2015

INTRODUCTION

Coal is the largest energy resource in Indonesia followed by natural gas, crude oil, hydro power and geothermal, respectively. However, oil and natural gas are currently still the main primary energy sources in the country and this pattern has to be changed due to the decreasing reserves of oil. The Government's energy diversification policy encourages using coal as a substitute for oil and gas for major domestic industries (Anonymous, 2013).

Due to the developing industry and growing population in Indonesia, electricity consumption and therefore demand for electricity generation has increased. Power sector now absorbs about 65% of domestic coal consumption and the figure is expected to increase in the future.

According to the Directorate General of Mineral and Coal data, the domestic demand on coal increases by an average of 5% per year from 2010 to 2013, mostly by the PLN (National Electricity Enterprise), independent power producers (IPPs), and captive power plants (i.e. PT Freeport Indonesia, PT Newmont Nusa Tenggara) while increasing demand on electricity is projected to reach amount of 9.4% per year (Anonymous, 2012).

This increasing trend in the electricity demand leads construction of new thermal power plants. However, environmental problems of fly ash such as wide area requirements for disposal, pollution of air, soil and water associated with ponds of ashes stored near the power plants have become preventing items for coal-fired power plants (Baykal and Saygili, 2011).

The utilization of coal is limited by the presence of high levels of ash and sulfur in it. During coal combustion, the mineral matter transforms into ash, and the amount of ash is so large and it is not easy to utilize ash effectively (Tamura, 2010). Fly-ash is the finely divided mineral residue resulting from the combustion of ground or powdered coal in electricity generating plant. It consists of inorganic matter present in the coal that has been fused during coal combustion (Pandey, et al., 2011).

Mineral matter in coal causes several disadvantages, including: unnecessary cost for transportation, handling difficulties during coal processing, leaching of toxic elements during ash disposal, sulfur emission from pyrite-like minerals giving rise to an environmental problem, and ash deposition leading to the deterioration of boilers and accessories (Ambedkar, 2012).

Currently, ash content reducing from the coal through coal washing is very common as a part of efforts to improve the quality of coal. The conventional methods such as gravity or froth flotation can produce coal with ash content is about 5 to 8%. Recently, thermal extraction of coals with organic solvents has been used to produce ashless coal which can be fired directly in gas turbines to achieve a higher net power output, resulting in reduced CO_2 emissions (Li, et al., 2004 and Baba et al., 2010).

In the coal washing, the first thing to be considered is washing method that will be applied to prepare the coal demand purposes, and whether washing is necessary because in principle coal can be sold immediately after the coal is mined. In fact, the direct sales of coal after mined means a producer does not get the maximum benefit. Therefore, in deciding a commercial considerations, coal washing should be included (Campbell, et al., 2010).

On the assumption that the coal needs to be processed with maximum benefit and meet the desired quality criteria of ash, water and sulfur contents, coal in size >0.5 mm is processed by gravity concentration. The coal with sized of <0.5 mm needs be studied carefully wether the coal needs to be processed or not, since the capital and operating costs of fine coal processing are very high, 4 to 5 times greater than the coarse coal processing. If the fine coal is necessary to be processed, then the most appropriate method would be froth flotation. The washing method based on coal particle size can be seen in Figure 1 (Wittmers, 2011).

Coal de-ashing technology can also be applied to optimize the yields of coal mining that is currently abandoned or not mined, because the position of the coal at the roof or bottom layer (Figure 2) usually has high ash content due to contamination of overburden (Schwerdtfeger, 2010).

In this work, in order to reduce the ash content for producing ash free coal (AFC), Peranap coals was treated under different solvents namely 1-methyl naphthalene ((Lee, 2012; Okuyama, et al., 2005), 1-1-1-methoxy ethoxy acetic acid and N-methyl 2 pyrolidynon (Li, et al., 2004). The effect of solvent

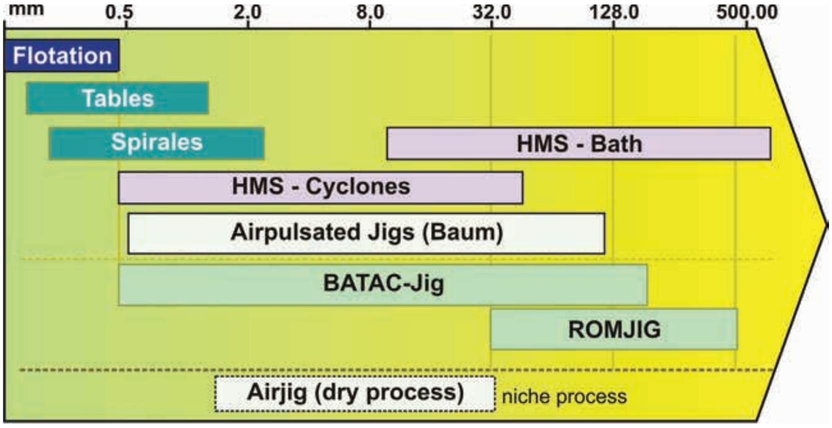

Figure 1. Methods of coal washing based on coal particle size

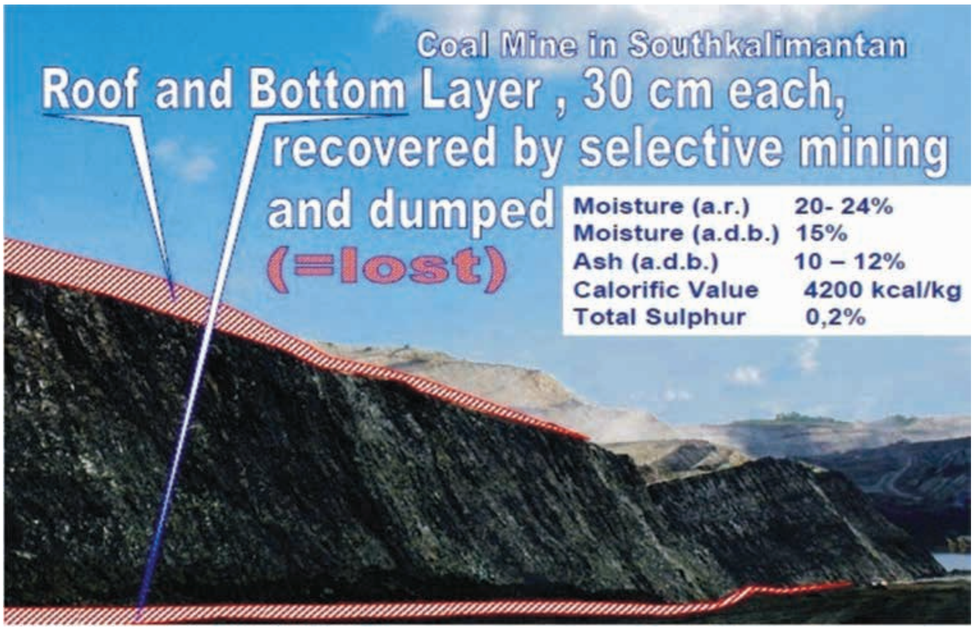

Figure 2. Roof and bottom coal layers

types and process conditions on reducing the ash content for two kinds of coal is presented and discussed in this paper.

METHODOLOGY

Coal de-ashing was carried out to obtain coal with very low ash content (ash free coal). The research activities were conducted through the following stages:

- Coal sampling, in Peranap coal mine from mine site and stockpile,
- Coal preparation, crushing, grinding and sizing,
- Coal characterization, proximate, ultimate and calorific value,
- Coal separation by using solvent,
- Coal de-ashing process experimentals,
- Ash content analysis of the products, and
- Evaluation

Coal samples for this research were taken from Peranap coal mining area in Riau province. There were two coal sampling locations, mine site and stockpile. In laboratory, the samples were dried, crushed, ground and sized to get the coal particle size of less then 65 mesh for coal characterization and less then of 8 mesh for coal separation through sink and float tests.

The characterization of the coals was conducted through proximate analysis that consists of inherent moisture, ash and volatile matter contents; ultimate analysis that consists of carbon, hydrogen, nitrogen and sulphur contents and calorific value of those coals. The results of coals analysis can be seen in Table 1.

Table 1. The analysis result of Peranap coals

Analysis	Mine site	Stockpile
Total moisture, % ar	38.44	29.32
Inherent moisture, % adb	21.58	19.32
Ash, % adb	2.65	5.65
Volatile matter, % adb	38.69	38.82
Fixed carbon, % adb	37.08	36.21
Carbon, % adb	52.55	50.2
Hydrogen, % adb	6.57	6.35
Nitrogen, % adb	0.79	0.67
Sulphur, % adb	0.22	0.34
Oxygen, % adb	37.22	36.79
Calorific value, cal/g adb	4,704	4,668

The coal separation to obtain coal with low and high ash contents was carried out by using per chlor ethylene (PCE) solvent with relative density of 1.6. The coal with particle size of less than 8 mesh was immersed in the liquid then the floating fraction being skimmed off and weighed, while the sinking fraction was separated and weighed. The denser particles, represent material with high ash content (Carillo, et al., 2012).

Coal de-ashing process is an ash-free coal manufacturing process applying solvent de-ashing technology. The high ash coal is extracted into coal derivative two ring aromatics at temperature of 300-350°C (Okuyama, et al., 2005). The solution is separated from the insoluble coal and ash by filtration. The solvent is recovered and recycled in the process. Ash-free extracted coal, known as ash free coal (AFC) and ash-concentrated insoluble coal (insoluble), are obtained in this process. The product is analysed for ash content.

Three kinds of solvent were tested namely 1-methyl naphtalene (MN), 1-1-1 methoxcy ethoxcy acetic acid (MEAA) and N methyl 2- pyrolidynon (MP). The use of MEAA and MN, the reaction time and processing temperature were kept constant i.e. for one hour at temperature of 300°C (Yoshida, et al., 2002). While using MP, the reaction time was tested in 30, 60 and 90 minutes at temperature of 300°C. The flow diagram of de-ashing process is shown in Figure 3.

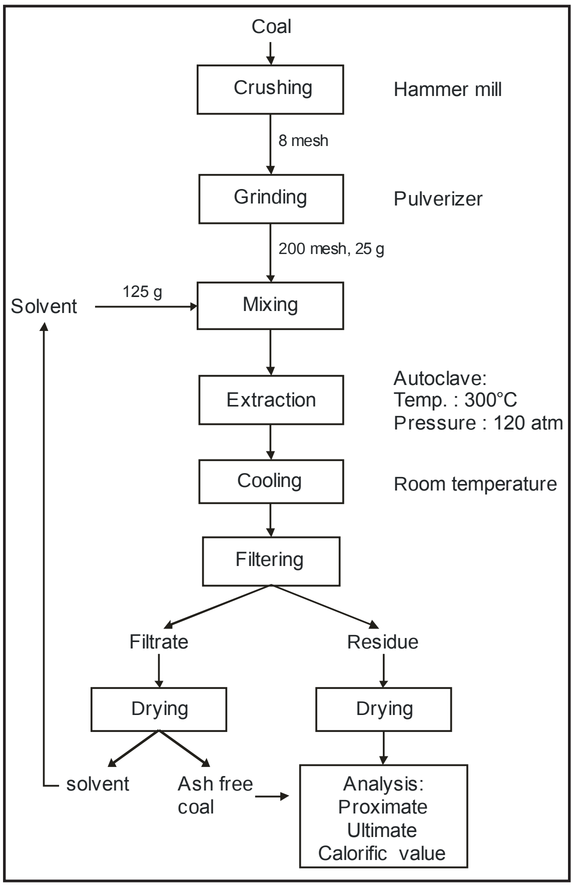

Figure 3. Flow diagram of coal de-ashing process

RESULTS AND DISCUSSION

The coal samples from stockpile and mine site after preparation and separation, were then extracted. The first experiment was carried out by using methoxy ethoxy acetic acid (MEEAA) and was heated at a temperature of 300°C for 1 hour. The ratio of coal and solvent was 1:6 w/w. The result of the experiment is shown in Table 2.

As can be seen in Table 2, the ash content of the extracted coal (AFC) from stockpile coal decreased from 46.02 to 14.44% for the first experiment and to 16.54% for the second experiment, while the AFC from mine site coal, the ash content decreased from 25.43 to 7.2% and 7.79% for the first and second experiments respectively. The

Table 2. Ash content after solvent extraction using MEEAA

MEEAA	Ash content. % adb			
	Stockpile		Mine site	
	Experiment 1	Experiment 2	Experiment 1	Experiment 2
Parent coal	46.02	46.02	25.43	25.43
Extracted coal (AFC)	14.44	16.54	7.2	7.79
Residual coal	57.84	57.60	22.08	32.03

ash content of residual coal increased from 46.02 to 57.84 and 57.60% for the stockpile's coal and for the mine site's coal. The ash content in the residual coal decreased in the first experiment but increased in the second experiment. According to Lee (2012), as can be seen in Table 3, the decreasing ash content in the extracted coal caused the increasing of ash content in the residual coal. Therefore, the result of this experiment is in line with Lee's experiment.

Table 3. Analysis results of advance coal cleaning (ACC) process

Sample	Proximate analysis (wt%)				Ultimate analysis (wt%, daf*)					
	Moisture	Volatile matter (daf)	Ash (dry)	Fixed carbon (daf)	C	H	N	O	BET (m^2/g)	Calorific value (kcal/kg)
Parent coal (PC)	15.4	39.0	4.6	41.0	69.0	4.3	0.3	26.4	3.9	5,450
Extracted coal (EC)	-	33.6	<0.01	66.4	87.9	4.3	0.0	7.9	0.2	8,560
Residual coal (RC)	-	32.5	8.0	59.5	87.7	4.2	0.0	8.0	4.1	7,490

Tabel 4. Ash content after solvent extraction using MN

MN	Ash content. %			
	Stockpile		Mine site	
	Experiment 1	Experiment 2	Experiment 1	Experiment 2
Parent coal	46.02	46.02	25.43	25.43
Extracted coal (AFC)	0.06	0.36	0.11	0.19
Residual coal	17.59	20.26	25.08	26.06

MEEAA ($CH_3(OCH_2CH_2)_2OCH_2CO_2H$) is an organic compound, categorized as a heterocyclic compound. This solvent is characterized as dispersant to improve the separation of particles and to prevent settling or clumping. The use of MEEAA to reduce the ash content in coal was effective because the ash content decreased significantly. However, the aim of this research is to obtain the coal with a very low ash content, therefore the use of MEEAA alone was not appropriate. The use of MEEAA should be together with another solution that has a function as mineral matters solvent (Li et al., 2004).

By using 1-methyl naphtalene (MN) as a solvent, coal de-ashing process has been conducted at a temperature of 300°C for 1 hour. The ratio of coal and solvent was 1:6 w/w. The results of the experiment can be seen in Table 4.

Coal de-ashing by using MN solution was very effective to reduce the ash content in the extracted coal. The ash content decreased significantly from 46.02 to 0.06 and 0.36% for the stockpile's coal and to 0.11 and 0.19% for the mine site's coal. By using the MN solution, the ash content reduced not only for the extracted coal, but also for the residual coal from stockpile's coal. The ash content of residual coal decreased from 46.02 to 17.59 and 20.26%. While the ash content of residual coal from mine site relatively unchanged. This phenomenon might be caused by the differences of impurities that contained in the stockpile and mine site coals.

By using N-methyl 2 pirolidinon with a ratio of coal and solvent of 1:5 w/w, the process was conducted at temperature of 300°C for 30, 60 and 90 minutes. The effect of residence time on coal ash content can be seen in Figure 4 and 5.

From Figure 4, it can be seen that the lowest ash content of the extracted coal from stockpile can be reached at the residence time of 30 minutes (ash content 2.68%). The more the time, the higher the ash content of the residual coal but relatively unchanged for the extracted coal. Different from the stockpile's coal, the mine site's coal produced an extracted coal with the lowest ash content at the processing time of 60 minutes (3.04%). In this case, both the extracted and residual coals, the ash content can be decreased. The ash contents

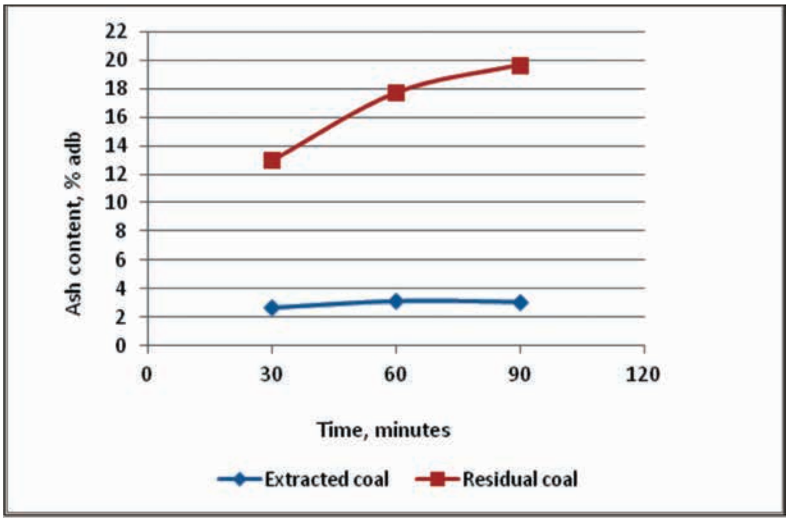

Figure 4. Effect of residence time on coal ash content (stockpile's coal)

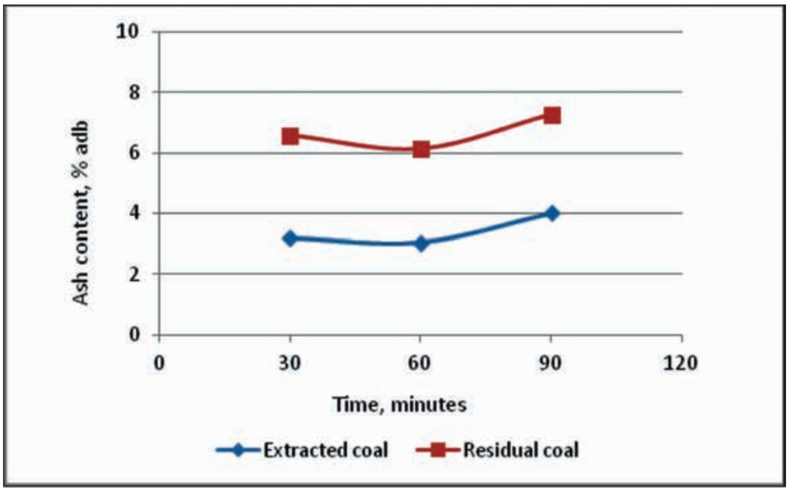

Figure 5. Effect of residence time on coal ash content (mine site's coal)

of the parent coals in this experiment (stockpile and mine site coals) were 21.49 and 8.23% respectively.

CONCLUSION

The experimental results indicate that the use of 1-methyl naphtalene (MN) was very effective to produce ash free coal (AFC) compared to 1-1-1 methoxcy ethoxcy acetic acid (MEAA) and N methyl 2- pyrolidynon (MP). The ash contents of the AFC by using Peranap coal as parent coal were 0.06% and 0.11 % in air dried basis (adb) for stockpile and mine site coals respectively. Those results were obtained at process temperature of 300°C, residence time of 1 hour and the ratio of coal and solvent 1:6 (w/w).

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the head of R&D Center for Mineral and Coal Technology (tekMIRA) for making this research possible. The authors also gratefully acknowledge PT Tambang Batubara Bukit Asam (PTBA) for full support during coal sampling. Thanks also go to the staffs of coal laboratory of tekMIRA for their full assistance during laboratory works.

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