THE BEHAVIOR OF HEAVY METALS CONTENT IN COAL COMBUSTION PRODUCTS (CCPS) AND ITS LEACHATE FROM INDONESIA COAL POWER PLANTS

SIFAT KANDUNGAN LOGAM BERAT PADA LIMBAH PEMBAKARAN BATUBARA DAN AIR LINDIANNYA DARI PEMBANGKIT LISTRIK TENAGA UAP (PLTU) DI INDONESIA

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ABSTRACT

The development of many coal power plants in Indonesia has been creating Coal Combustion Products (CCPs) in a huge amount. The generating coal power plant will increase dramatically from 50 to 320 TWh in 2020. It is predicted that the total CCPs will be nearly 10.8 million tons in the same year. The large quantity of Indonesia CCPs will likely increase drastically and potentially will be a serious problem in the future. This research aims to measure heavy metals content in coal and CCPs, to assess their distribution in leachate and investigate the concentration level of heavy metals in leachate using TCLP method, and also to analyze the correlation between heavy metals content in coal, CCPs, and CCPs leachate using Pearson analysis.

The analysis results show that the dominant element content in coal was boron. Moreover, the distribution of heavy metals tended to enrich fly ash. The concentration level of heavy metals fly ash and bottom ash leachates from all the power plants generally was much lower than the standard threshold. The significant level of concentration on fly ash and bottom ash was shown by boron. The concentration levels of heavy metals of coal ash leachates from two power plants were also much lower than the standard limit. The correlation between the heavy metals content of parent coal and CCPs pointed to no correlation between the variables. The heavy metals content of coal had no correlation with the concentration of heavy metals in CCPs leachate excluding nickel and chromium in bottom ash. Finally, it is recommended to assess other heavy metals concentration such as arsenic, mangan and selenium in CCPs leachate and further conduct a long-term study about the characteristics, leaching behavior of heavy metal leachate and, their effects on the environment.

Keywords: heavy metals, coal combustion products, leachate, heavy metals distribution

SARI

Pembangunan PLTU berbahan bakar batubara di Indonesia, telah menghasilkan limbah hasil pembakaran batubara dalam jumlah besar. Pada 2020, jumlah pembangkit listrik berbahan bakar batubara akan meningkat dari 50 menjadi 320 TWh. Diperkirakan jumlah limbah pembakaran batubara yang dihasilkan dalam setahun sebanyak 10.8 juta ton. Jumlah limbah batubara yang sedemikian besar akan menimbulkan dampak negatif terhadap lingkungan. Penelitian ini bertujuan untuk mengukur kandungan logam berat pada batubara dan limbah pembakaran batubara, mengetahui kandungan dan distribusi logam berat pada air lindian dengan metode TCLP dan menganalisis hubungan kandungan logam berat pada batubara, limbah pembakaran batubara dan

Received : 09 January 2014, first revision : 24 January 2014, second revision : 11 June 2014, accepted : June 2014

air lindiannya menggunakan analisis Pearson. Hasil analisis menunjukkan bahwa rata-rata kandungan logam berat pada batubara didominasi oleh boron. Selanjutnya, distribusi logam-logam berat yang ada cenderung untuk menumpuk pada abu terbang. Konsentrasi logam berat pada abu terbang dan abu dasar di semua lokasi PLTU diketahui masih berada jauh di bawah ambang batas yang telah ditetapkan dan didominasi oleh unsur boron. Hasil perhitungan mengenai hubungan antara kandungan logam berat pada batubara dan limbah pembakaran batubara menunjukkan tidak terdapat hubungan nyata. Kandungan logam berat yang terdapat pada batubara tidak berkaitan dengan konsentrasi logam berat pada air lindian limbah pembakaran batubara, kecuali yang terdapat nikel dan kromium pada abu dasar. Penelitian ini merekomendasikan beberapa masukan di antaranya untuk dilakukan pengukuran lanjutan pada beberapa logam berat yang lain seperti arsenik, selenium dan mangan. Direkomendasikan pula dilakukan kajian jangka panjang terkait dengan karakteristik dan perilaku logam berat pada air lindian dan pengaruhnya terhadap lingkungan.

Kata kunci : logam berat, limbah pembakaran batubara, air lindian, distribusi logam berat

INTRODUCTION

Coal is an important material in the Indonesian energy sector. It is widely utilized in the provision of energy supply in Indonesia. In 2011, total installed capacity of coal power plant was about 10,177 MW and total consumption of coal was 27.4 million tons (Directorate General of Electricity, 2011). The development of coal power plants in Indonesia has produced coal combustion products (CCPs) in a huge amount. A coal power plant generally produces about 8-10% of CCPs. Thus, the CCPs produced in that year were about 2.7 million tons. The generating coal power plants will increase dramatically from 50 to 320 TWh in 2020 and the supply of coal will be 108.3 million tons per year (Anonymous, 2006). It is predicted that the total CCPs will be nearly 10.8 million tons in the same year. A large quantity of CCPs will potentially be a serious problem in the future owing to the requirements for storage. During transportation disposal and storage phases; the residues from CCPs are subjected to the leaching effects of rain. A portion of their undesirable components found in ashes can pollute both ground and surface water (Benito et. al,. 2001). Consequently, additional environmental problems could emerge. The objectives of this study are to measure the heavy metals content in coal, CCPs and CCPs leachate using TCLP method; as well as to analyze their correlations using Pearson analysis.

METHODOLOGY

Samples of coal, fly, bottom and coal ashes were obtained from nine coal power plants in Indonesia. Coal ash is a mixture of fly and bottom ashes. The type and code of the samples are shown in Table 1. A direct acid digestion method using a microwave reaction system was carried out to determine heavy metal content. About 0.5 gram of dry fly, bottom and coal ashes was selected from the composite samples and weighed. Later, 0.1 gram of coal was used for this step. The sample was digested with 2.5 ml of HNO_3 and 7.5 ml of (HCl). The digested material was then filtered and diluted to 50 ml with distilled water. On the other side of the process, the TCLP procedure was based on EPA Method 1311. Using this method, the CCPs samples were subjected to 18 ± 2 hours with the leaching solution. The leaching solution was a mixture of CH_3CH_2OOH (glacial acetic acid), reagent water and 1N NaOH. The solution was diluted to a volume of 1 liter to have a pH of 4.93 ± 0.05. Then, the extract samples were analyzed by an ICPS-8100 Sequential Plasma Spectrometer to determine the level of heavy metal content (US EPA, 1992).

In order to measure the degree of relationship between samples, the correlation will represent its relationship. The correlation coefficient communicates both the strength and the direction of association that it has index number between the range of -1 and +1. The negative numbers representing a negative correlation which means as one variable increases so the other variable decreases. On the other hand, positive numbers representing a positive correlation which means since one variable increases, so does the other.

A correlation matrix is presented to describe whether one or more variables have an association. Then, the effect size is used to assess the importance of an effect between two sets of data. Generally, the distribution level of correlation is devided into three level's (Santoso, 2012):

- r = 0.0 – 0.5 has weak correlation.
- r = 0.5 – 0.7 has strong correlation.
- r = more than 0.7 has very strong correlation.

Table 1. Type and code of research samples.

No.	Power plant	Type of Sample	Sample Code
1	Ombilin	Coal Coal Ash	CA CaA
2	Tanjung Enim	Coal Coal Ash	CB CaB
3	Tanjung Jati Unit 1	Coal Fly Ash Bottom Ash	CC FC BC
4	Tanjung Jati Unit 2	Coal Fly Ash Bottom Ash	CD FD BD
5	Tanjung Jati Unit 3	Coal Fly Ash Bottom Ash	CE FE BE
6	Tanjung Jati Unit 4	Coal Fly Ash Bottom Ash	CF FF BF
7	Rembang	Coal Fly Ash Bottom Ash	CG FG BG
8	Paiton I	Coal Fly Ash Bottom Ash	CH FH BH
9	Paiton 9	Coal Fly Ash Bottom Ash	CI FI BI

RESULTS AND DISCUSSION

Heavy Metals Content of Coal

The heavy metals content of coal is shown in Table 2. It can be seen that the dominant heavy metals content in the coal was boron. The content of boron in CB, CC, CD, CE, and CF samples were 151, 175, 94.7, 101 and 94.9 mg/kg respectively The average content of boron was 87 mg/kg, followed by barium (64.4 mg/kg). Meanwhile, the average content of cadmium (0.8 mg/kg) was the lowest one. Iyer (2002) stated that the characteristics of the coal had direct influence on the chemical and mineralogical composition of the CCPs.

In terms of Indonesian coal, the overwhelming majority either lignite or sub-bituminous comprises 86.61 % of Indonesian coal (Suhala, 2011). The power plant in this study utilized two types of coal rank, namely lignite and sub bituminous coal rank. The means value of heavy metals content for both coal ranks is shown in Table 3.

According to Table 3, it was clear that boron and barium in both coal ranks were dominant and several times higher than that of the other elements. The lowest content was cadmium; in sub bituminous was 0.85 mg/kg and 0.77 mg/kg in lignite coal rank.

Distribution of Heavy Metals in Fly Ash and Bottom Ash

The distribution between the bottom and fly ash fraction is a function of the coal and the boiler types. In dry-bottom boilers, fly ash constitutes the major ash component at 80–90 % with bottom ash in the range of 10–20 %. Wet-bottom boilers produce slag, from the furnace bottom (Departement of Environment USA, 2006). Many of the most toxic elements, significant enrichment is observed in the fine particle of fly ash (Nelson, 2010).

Figure 1 shows barium distribution in fly and bottom ashes. Five data were above the line while two of them were on the opposite side. Those data indicated that barium tends to enrich the fly ash. The element in fly ash forms sparingly soluble compounds with carbonates and sulphates. Particular attention should be given to Ba metalates because its low solubility would also attenuate Ba releases (Cornelis et al., 2008)

Figure 2 reveals chromium distribution in fly and bottom ashes. It is clear that the position of all data was above the line. It means that chromium tends to enrich the fly ash. Chromium in its hexavalent oxidation state is widely recognized as potentially carcinogenic and highly soluble in aqueous media (Huggins and Huffman, 2004).

Figure 3 presents the distribution of cobalt in bottom ash and fly ash. Five cobalt data appeared on the upper side of the line while two data were under the line, so cobalt tends to enrich the fly ash.

Figure 4 shows nickel distribution in fly and bottom ashes. Nickel also tends to enrich the fly ash.

Figure 5 depicts copper distribution fly and bottom ashes. Copper tends to enrich the fly ash and, is assimilated within the glass not easily released. In contrast, the oxidation of Cu-Fe sulphides in coal leads to a higher mobility of Cu in fly ash (Soco and Kalembkiewicz, 2007).

Figure 6 depicts zinc distribution distribution zinc in fly and bottom ashes. The element tends to

Table 2. Heavy metals content in coal sample (mg/kg)

Sample Code	Content (mg/kg)								
	Ba	Cr	Co	Ni	Cu	Zn	Cd	B	Pb
CA	46.0	9.2	4.7	8.8	12.6	25.6	1.2	41.2	65.2
CV (%)	0.3	1.2	2.2	0.3	1.2	5.1	2.0	1.2	71.6
CB	50.5	2.1	2.8	2.4	3.0	15.4	0.5	151	13.6
CV (%)	0.0	12.8	2.7	1.3	0.1	10.4	9.8	2.0	22.1
CC	120	12.6	4.7	9.4	5.4	16.5	1.2	175	43.8
CV (%)	60.2	53.5	35.6	36.9	41.4	36.8	55.4	55.8	61.6
CD	59.2	5.5	3.2	6.1	2.9	15.0	0.6	94.7	11.7
CV (%)	0.3	5.0	1.9	4.4	23.8	1.0	6.4	0.3	3.5
CE	72.4	6.9	2.7	5.8	4.4	38.1	0.7	101	10.6
CV (%)	1.7	1.0	3.6	2.3	15.7	3.0	11.1	1.7	23.4
CF	79.1	9.6	3.3	7.1	4.1	44.0	0.9	94.9	13.8
CV (%)	3.1	4.6	2.7	4.1	4.0	0.4	16.1	0.4	20.0
CG	62.5	2.7	3.7	5.4	2.3	11.3	0.7	62.4	6.6
CV (%)	0.1	1.1	2.0	2.7	0.1	2.7	4.7	0.7	9.0
CH	37.6	0.7	2.1	2.9	1.5	2.7	0.4	32.1	2.1
CV (%)	20.7	11.5	15.4	13.4	3.7	12.0	11.7	19.8	8.7
CI	52.5	2.4	5.3	5.4	2.2	8.5	1.2	30.9	5.9
CV (%)	40.3	17.2	0.7	0.3	6.7	9.7	2.1	1.3	28.9
Average	64.4	5.8	3.6	5.9	4.2	19.6	0.8	87.0	19.3

Table 3. Mean value of heavy metals content based on coal rank

Elements	Sub Bituminous (mg/kg)		Lignite (mg/kg)	
	Mean	Standard Deviation	Mean	Standard Deviation
Ba	71.10	26.80	50.90	12.50
Cr	7.66	3.66	1.93	1.10
Co	3.54	0.93	3.68	1.60
Ni	6.58	2.52	4.59	1.44
Cu	5.39	3.65	1.97	0.43
Zn	25.70	12.60	7.47	4.38
Cd	0.85	0.30	0.77	0.40
B	110.00	47.20	41.80	17.90
Pb	26.50	22.80	4.89	2.42

enrich into the fly ash.

Figure 8 reveals boron distribution in fly and bottom ashes. There is one piece data along the line but six points on the upper side of the line. Generally, it is found that boron tends to enrich the fly ash.

Figure 9 depicts lead distribution distribution lead fly- and bottom ashes . Lead also tends to enrich fly ash. Around 50-60% Pb is estimated to be in surface association with fly ash (Spears and Martinez-Tarrazona, 2004).

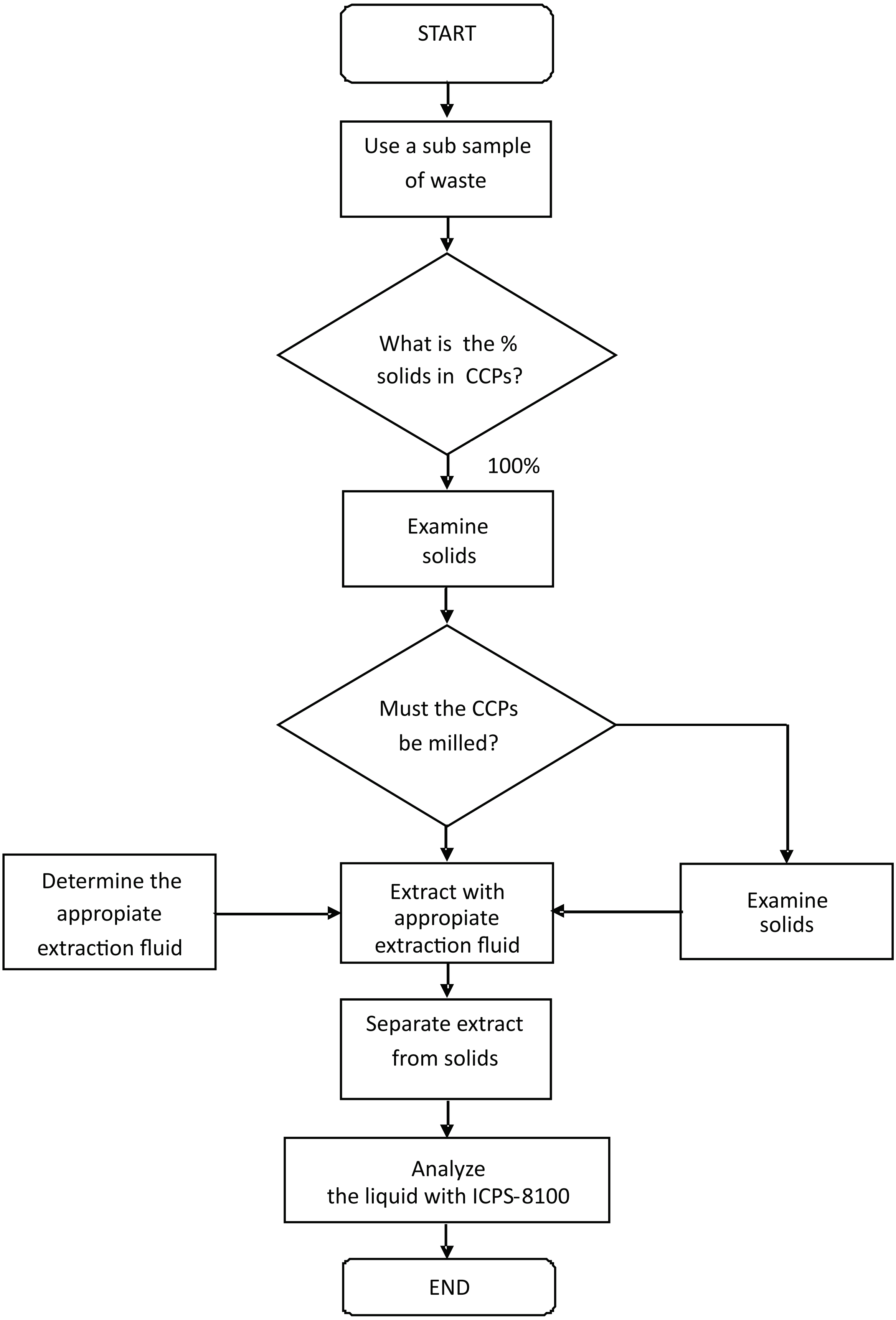

Figure 1. Flowchart of Toxicity Characteristic Leaching Procedure (TCLP) EPA Method 1311

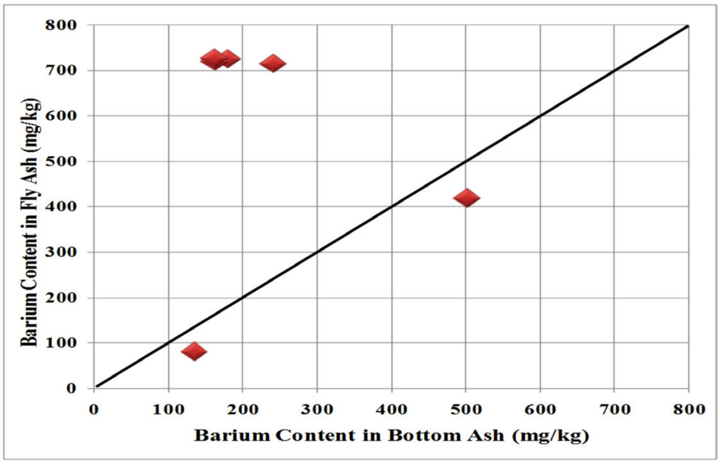

Figure 1. Distribution of barium in fly and bottom ashes

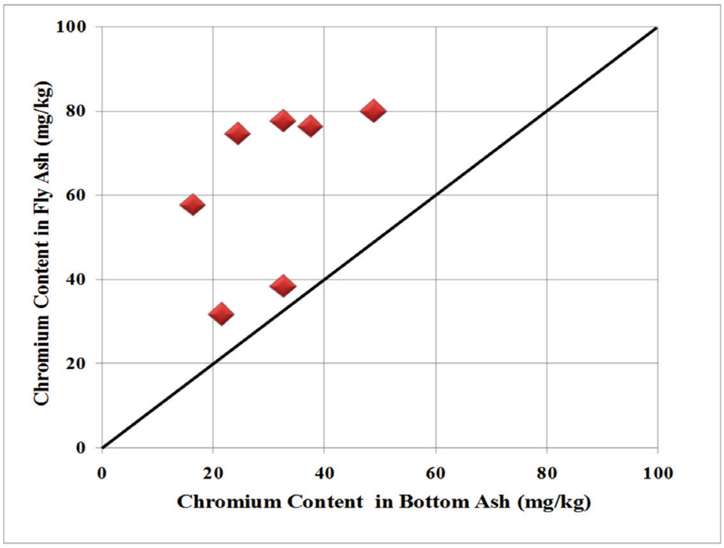

Figure 2. Distribution of chromium in fly and bottom ashes

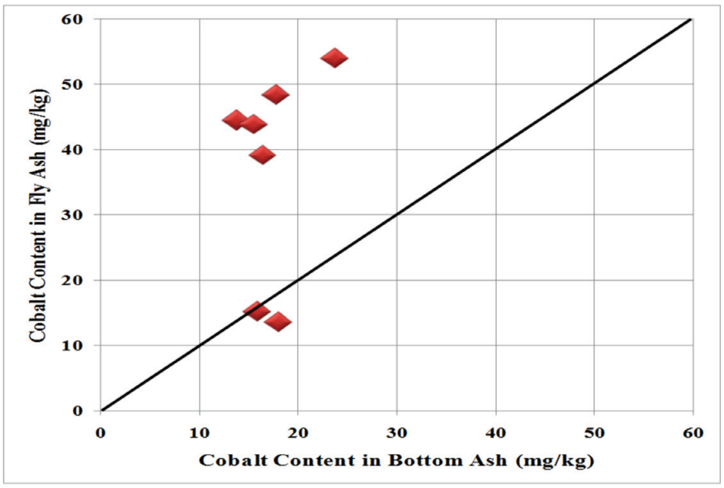

Figure 3. Distribution of cobalt in fly and bottom ashes

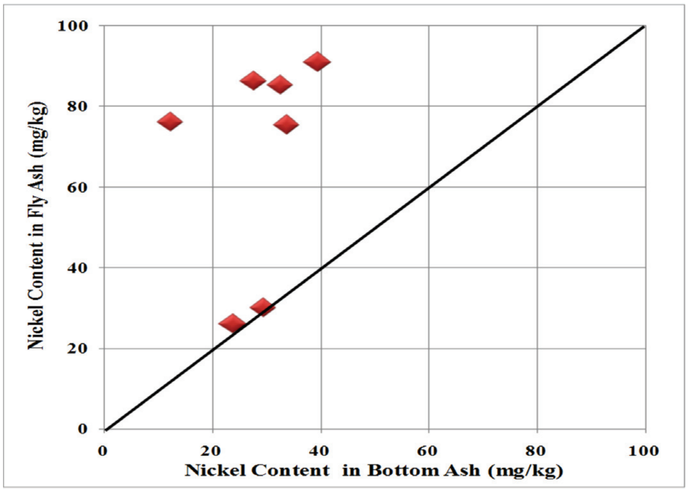

Figure 4. Distribution of nickel in fly and bottom ashes

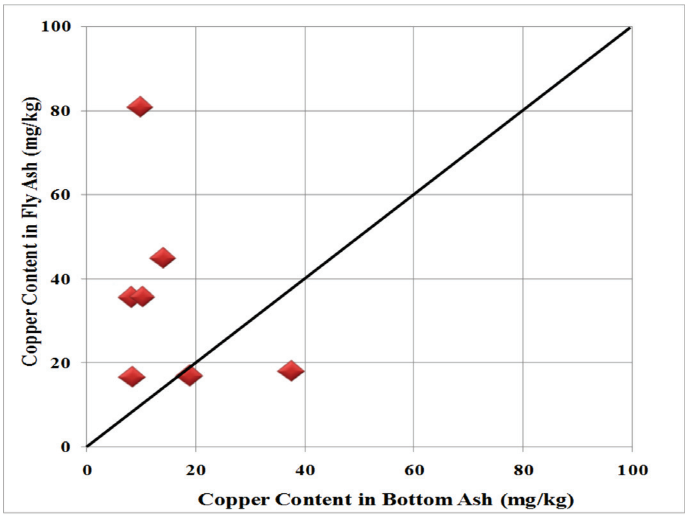

Figure 5. Distribution of copper in fly and bottom ashes

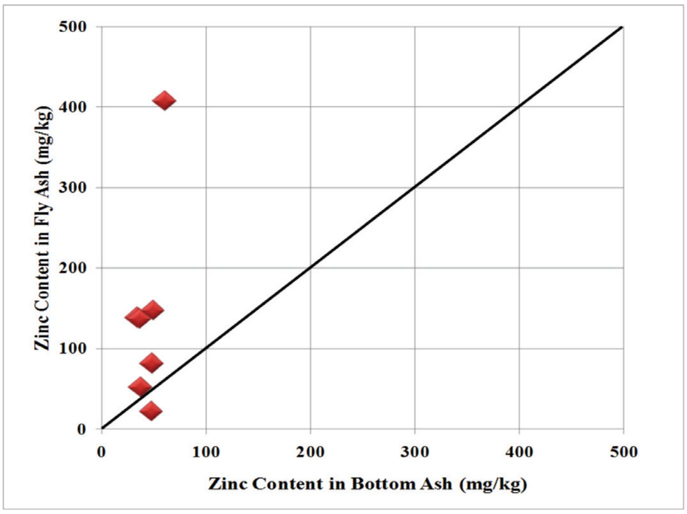

Figure 6. Distribution of zinc in fly and bottom ashes

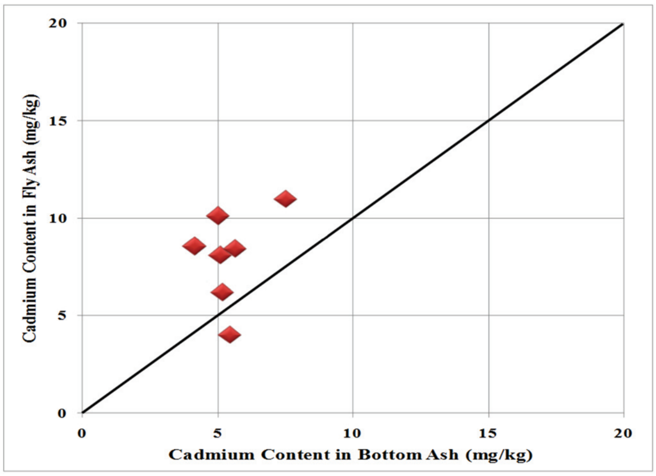

Figure 7. Distribution of cadmium in fly and bottom ashes

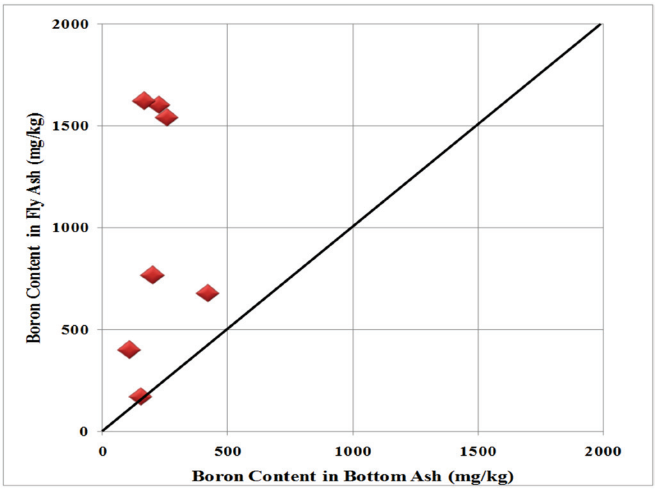

Figure 8. Distribution of boron in fly and bottom ashes

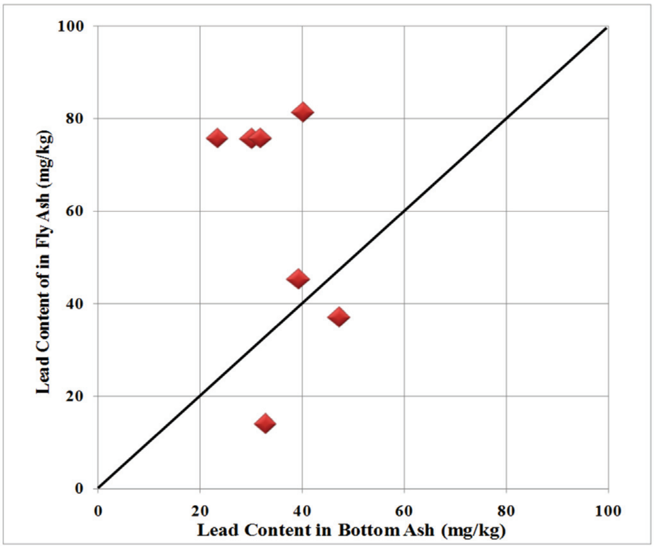

Figure 9. Distribution of lead in fly- and bottom ashes

Based on the data, all heavy metals tend to enrich the fly ash as a result of elements volatilization in the boiler and their subsequent condensation in the cooler sections of the flue gas stream. Karayigit et.al., (2005) indicated that some of the volatile elements notably As, Cd and Zn increase from a coarse to a finer particle size of fly ash. Volatile elements like Zn and As will increase in concentration as a function of their decreasing particle size and consequently enhanced surface area of the fly ash. The content of volatile trace elements thus will increase with an increase in fly ash surface area (Hower et al., 2001).

Concentration of Heavy Metals in Leachate (TCLP)

The concentration of heavy metals in fly ash leachates is shown in Table 4. On average, the concentration of boron (22.4 mg/L) was many times higher than that other elements (under 1.0 mg/L). Cadmium is not found in any of the seven fly ash leachates. Interestingly, the amount of Zn observed in the FI leachate (4.84 mg/L) is higher than that of other fly ash samples. The concentration of volatile elements such as Zn will increase as a function of decreasing particle size and enhanced the surface area of fly ash. The fine particle fraction of fly ash is enriched by trace elements compared with the fraction of trace elements in the parent coal (Davison, 1974).

Heavy metals concentration in bottom ash leachate is shown in Table 5. Generally, the concentration level of heavy metals in bottom ash leachate is much lower if compared to standard limit. Concentration of boron in bottom ash leachate also dominates. Copper has the lowest concentration in the fly ash leachate. However, boron concentration concentration boron in bottom ash is less than that in fly ash. Boron concentration in the bottom ash leachate is considerably lower than that of boron in the fly ash.

The concentration level of heavy metals in the coal ash leachates is shown in Table 6. It can be seen that the concentration level from two power plants are lower and under standard limit. These concentrations for most heavy metals are under 1 mg/L except for boron (8.51 mg/L). To sum up, the concentration level of heavy metals in CCPs is far below the standard threshold.

Pearson Correlation Analysis

Table 7 shows Pearson correlation matrix for heavy metal content of coal, flyand bottom ashes, based on the accumulation of heavy metal content

Table 4. Concentration of heavy metals in fly ash leachates (mg/L)

Sample Code	Concentration (mg/L)								
	Ba	Cr	Co	Ni	Cu	Zn	Cd	B	Pb
FC	0.29	0.05	0.02	0.10	0.00	0.15	0.00	36.10	0.05
CV (%)	36.90	40.30	6.05	4.17	-	24.70	-	5.50	20.00
FD	0.20	0.04	0.02	0.11	0.00	0.14	0.00	37.40	0.05
CV (%)	36.60	37.60	0.50	1.33	-	18.50	-	0.45	8.12
FE	0.60	0.03	0.01	0.05	0.00	0.06	0.00	28.9	0.04
CV (%)	13.10	6.48	1.72	7.36	-	41.30	-	1.34	8.27
FF	0.75	0.00	0.00	0.03	0.00	0.04	0.00	20.40	0.02
CV (%)	6.83	66.20	55.3	3.20	-	12.20	-	0.18	14.20
FG	0.36	0.00	0.00	0.01	0.02	0.04	0.00	0.75	0.00
CV (%)	0.28	-	-	100.00	47.20	12.30	-	0.82	-
FH	0.37	0.08	0.00	0.03	0.00	0.00	0.00	20.70	0.03
CV (%)	13.60	0.27	-	21.50	-	100.00	-	2.99	0.38
FI	0.29	0.02	0.04	0.10	0.00	4.84	0.00	12.60	0.02
CV (%)	1.15	4.86	8.53	15.90	-	7.09	100.00	1.00	0.82
Average	0.41	0.03	0.01	0.06	0.00	0.75	0.00	22.39	0.03

Table 5. Heavy metal concentration in bottom ash leachate (mg/L)

Sample Code	Concentration (mg/L)								
	Ba	Cr	Co	Ni	Cu	Zn	Cd	B	Pb
BC	0.17	0.02	0.03	0.04	0.02	0.11	0.02	1.86	0.04
CV (%)	0.02	0.02	0.03	0.02	0.02	0.03	0.02	0.21	0.04
BD	0.24	0.03	0.05	0.06	0.03	0.08	0.04	1.79	0.07
CV (%)	0.00	0.01	0.01	0.01	0.01	0.01	0.01	0.09	0.01
BE	0.46	0.04	0.03	0.06	0.03	0.23	0.04	1.78	0.04
CV (%)	0.01	0.00	0.01	0.03	0.00	0.01	0.01	0.02	0.01
BF	0.47	0.03	0.03	0.06	0.02	0.07	0.04	1.35	0.03
CV (%)	0.01	0.00	0.00	0.03	0.00	0.01	0.01	0.12	0.01
BG	0.37	0.03	0.03	0.06	0.02	0.10	0.04	0.70	0.02
CV (%)	0.02	0.00	0.01	0.02	0.00	0.01	0.01	0.22	0.00
BH	0.46	0.06	0.04	0.06	0.02	0.06	0.04	1.74	0.06
CV (%)	0.01	0.02	0.01	0.01	0.00	0.01	0.01	0.06	0.00
BI	0.52	0.04	0.04	0.05	0.04	0.04	0.04	0.62	0.13
CV (%)	0.02	0.00	0.01	0.01	0.01	0.00	0.00	0.14	0.01
Average	0.38	0.04	0.04	0.06	0.03	0.10	0.04	1.41	0.06

Table 6. Heavy metal concentration in coal ash leachates (mg/L)

Sample Code	Concentration (mg/L)								
	Ba	Cr	Co	Ni	Cu	Zn	Cd	B	Pb
Ca A	0.30	0.05	0.04	0.06	0.06	0.10	0.04	3.38	0.10
CV (%)	2.10	11.20	12.20	8.21	9.19	2.66	12.80	3.35	8.51
CaB	0.77	0.04	0.03	0.04	0.04	0.05	0.03	13.60	0.08
CV (%)	0.64	11.80	15.00	14.20	24.10	14.00	18.50	6.60	27.40
Average	0.53	0.05	0.04	0.05	0.05	0.08	0.03	8.51	0.09

Table 7. Pearson correlation between heavy metals content in coal and CCPs for each elements (n=7)

	Ba		Cr		Co	
	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash
Pearson Correlation	-0.05	0.07	0.28	0.10	0.08	-0.30
Sig. (2-tailed)	0.92	0.88	0.54	0.84	0.87	0.51
	Ni		Cu		Zn	
	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash
Pearson Correlation	0.02	-0.003	0.496	-0.268	-0.31	-0.65
Sig. (2-tailed)	0.97	0.996	0.258	0.561	0.50	0.11
	Cd		B		Pb	
	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash
Pearson Correlation	-0.003	-0.48	0.655	0.487	0.325	-0.368
Sig. (2-tailed)	0.996	0.274	0.110	0.268	0.477	0.417

in every power plant. In general, it is clear that no correlations of heavy metals content in coal, fly and bottom ashes, as evidenced by the significant correlation (p) result of fly ash and bottom ash that reveals more than 0.05. The range of significant (2-tailed) is 0.110 to 1.00. The level all of those elements is far above the level of p which means that there is no relationship between the element content of the coal to the element content of the fly ash or bottom ash . To identify the importance of size effect, Pearson correlation on fly and bottom ashes are r (7): -0.003 – 0.655 and -0.65 – 0.485 respectively. These correlations could mostly be categorized as weak correlation due to the number is being less than 0.5. But, because there is no correlation on variables, this number is meaningless.

Result of Pearson correlation for heavy metals content in coal and leachate concentration CCPs by TCLP is shown in Table 8. Significant correlation for the variables in fly ash reveals a range from 0.120 to 0.84 and in bottom ash is 0.01 to 0.809. The level of p is determined under 5 % ($p < 0.05$). Thus, there is was no correlation for the majority of the elements. However, for nickel and chromium elements, there was correlation due to $p < 0.05$; p for nickel and chromium; are 0.01 and 0.03 respectively. Moreover, the Pearson correlation result for nickel and chromium was -0.881 and -0.80, respectively, thus categorized as a very strong correlation between variable. It means when there is an increase in the number of concentrations of leachate in coal, the effect will be at the decreasing concentration for the CCPs leachate.

Table 8. Pearson correlation for heavy metals content in coal and the leachate concentration of CCPs by TCLP (n=7)

	Ba		Cr		Co	
	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash
Pearson Correlation	0.09	-0.63	-0.17	-0.80*	0.38	-0.24
Sig. (2-tailed)	0.84	0.13	0.71	0.03	0.40	0.60
	Ni		Cu		Zn	
	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash
Pearson Correlation	0.42	-0.881*	-0.047	-0.113	-0.28	0.52
Sig. (2-tailed)	0.36	0.01	0.286	0.809	0.54	0.23
	Cd		B		Pb	
	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash	Fly Ash	Bottom Ash
Pearson Correlation	a	a	0.643	0.550	0.519	-0.276
Sig. (2-tailed)			0.120	0.201	0.233	0.549

* : correlation significant at the 0.05 level (2-tailed)
a : cannot be computed because at least one of the variable is constant

CONCLUSIONS

It can be concluded that chemical characteristics of Indonesian coal are dominated by boron, Cadmium is only a little of that content. The heavy metals concentration level of CCPs leachates from nine power plants were far below the standard limit, so the CCPs leachates can be categorized as non-hazardous material. The majority of the heavy metals did not have any correlation in terms of the heavy metals content in coal and the heavy metals concentration in CCPs leachates except for nickel and copper.

It is necessary to assess the concentrations of other heavy metals such as arsenic, mangan and selenium in the CCPs leachate and furthermore. Conducting a long-term study on the characteristics and leaching behavior of heavy metal leachate and their effects to the environment. is suggested.

ACKNOWLEDGEMENTS

The author would like to thank PLTU Bukit Asam, PLTU Ombilin, PLTU Rembang, PLTU Tanjung

Jati B Unit 1-4, PLTU Paiton Unit 1 for providing the samples. Thanks also to Prof. Yutaka Dote of the University of Miyazaki for his supervision, support and valuable discussion.

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