THE DETERMINATION OF HEAVY METALS IN COAL ASH

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DECLARATION

No portion of work referred to in this dissertation has been submitted in support of an application for another degree or qualification of this or any other university or institution of higher learning.

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The Determination of Heavy Metals in Coal Ash

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ABSTRACT

Sejingkat Power Corporation Sdn. Bhd., is a coal-fired power which is located near Bako generates energy and electricity. Combustion of coals in coal-fired power plant generates huge amounts of ashes. When coal is burnt, it produces coal ash into the environment. Ash samples were collected from the ash pond in five different depths at eight different locations. The analyses of heavy metals were done in each depth of the different locations. The concentrations of heavy metals such as Cd, Mn, Zn, Pb and Ni in coal ash were analyzed using Atomic Absorption Spectrometry (AAS) method. Pb was found to have the highest concentration in the ash samples, followed by Zn and Ni. The mean concentrations of heavy metals were found to be in a following order in the ash samples: Mn < Cd < Ni < Zn < Pb. Among the five heavy metals, Pb is most abundant in the coal ash pond compared to Zn, Ni, Cd and Mn.
Keywords: heavy metal, coal, coal ash, coal-fired power plant, power plant

ABSTRAK

Sejingkat Power Corporation Sdn. Bhd. adalah merupakan stesen janakuasa elektrik yang menggunakan pembakaran arang batu untuk menghasilkan tenaga dan elektrik dan ia terletak berdekatan dengan Bako. Pembakaran arang batu dalam stesen janakuasa menghasilkan kuantiti abu yang banyak. Apabila batu arang dibakar, ia akan melepaskan abu ke alam sekitar. Sampel abu diperolehi dari kolam abu daripada lima kedalaman dan lapan lokasi yang berbeza. Analisis kandungan logam berat dijalankan untuk setiap kedalaman bagi lokasi yang berlainan. Kandungan logam berat seperti Cd, Mn, Zn, Pb dan Ni dianalisis dengan menggunakan Atomic Absorption Spectrometer (AAS). Pb didapati mempunyai kandungan yang tinggi dalam sampel abu diikuti oleh Zn dan Ni. Kandungan logam berat dalam sampel abu adalah: Mn < Cd < Ni < Zn < Pb. Antara lima logam berat tersebut, kandungan Pb adalah paling banyak berbanding dengan Zn, Ni, Cd dan Mn.
Kata kunci: logam berat, arang batu, abu, stesen janakuasa elektrik, kuasa elektrik

CHAPTER ONE

INTRODUCTION

Heavy metals are toxic to biota and the environment. Our environment has been increasingly polluted with various kinds of toxic pollutants and one of the pollutants is heavy metal which is found in various substances including coal.

Coal is used in power stations to generate energy to produce electricity. When coal is burnt, it produces coal ash. Heavy metals are also found in coal ash. Environmental pollution by thermal power plants based on coal all over the world is said to be one of the major sources of pollution affecting the general aesthetics of environment in terms of land use, health hazards and air, soil and water in particular and thus leads to environment dangers (Asokan *et al*., 2005). Previous research has shown that coal ash from the combustion of coal has an effect on the environment. If the ashes with heavy metals are released into the environment, they pollute the atmosphere, water and land. Consequently, they may become a hazard to the environment because of their contribution to the formation of toxic compounds (Baba and Kaya, 2003).

The Sejingkat Power Corporation Sdn. Bhd Coal-Fired Power Station at Kuching located by the Sarawak River adjacent to Kampung Goebilt in Muara Tebas, produces coal ash from the combustion of coal. Literature search has not yielded any study on heavy metals in the coal ash produced at this power plant. The objective of this study was

to determine the heavy metals in this coal ash and assess its potential for environmental pollution.

CHAPTER TWO

LITERATURE REVIEW

2.1 Heavy Metal

2.1.1 What is Heavy Metal?

Heavy metals are elements having an atomic weight between 63.546 and 200.590 which in their standard state, have a specific gravity (density) of more than about 5 g/cm^3 in their elemental form (Wild, 1996). Although many of the elements listed under 'heavy metals' have specific gravities greater than five, major exception to this rule remain. In hindsight, metals which have the specific gravities of more that five should preferably have been referred to as 'toxic elements', for they are all included in the United States Environmental Protection Agency's (USEPA's) (Sengupta, 2002). Metal being elements cannot be degraded nor created. They are an example of ultimate persistence (Samsiah, 2007). Examples of heavy metals are chromium (Cr), cadmium (Cd), lead (Pb), mercury (Hg) and manganese (Mn).

2.1.2 Environmental Effects of Heavy Metal

Human activity effects the natural geological and biological redistribution of heavy metals through pollution of the air, water and soil and also by altering the chemical form of heavy metals released in the environmental (Goyer, 1996; Hawkes, 1997). Mines, foundries, smelters and coal-burning power plants, as well as combustion by-products and vehicles emissions are the primary anthropogenic sources of heavy metals. When

present in high concentrations in the environment, heavy metals may enter the food chain from soils and result in becoming a health hazard (Mashi *et al.*, 2005). Heavy metals are the most common of all metabolic poisons. The mechanism of metals toxicity is different from other metabolic poisons. Metal toxicity can affect enzymes, the cellular proteins that react with and inhibit *sulfhydryl* (SH) *enzyme* systems, such as those involved in the production of cellular energy (Csuros and Csuros, 2002). Heavy metals toxicity tends to bioaccumulate in plants and animals, bioconcentrate in the food chain, or attack specific organs of the body (Goyer, 1997; Hawkes, 1997).

2.2 Heavy Metal in Coal

Generally, coal describes a large range of fossil fuels derived from partial degradation of plants and is a complex organic carbonaceous material with molecular structure which is not fully understood (Graham and Trotman, 1983). Weller *et al.* (1983) expressed coal as having 'an extensive macromolecular structure' or as 'possessing macromolecular networks containing trapped molecules' (Given *et al*., 1983a). According to Larsen (1985), modern scientists see coal as a 'macromolecular solid' or in another way 'as macromolecular organic rock' (Hatcher *et al*., 1983). Green *et al*. (1982) stated that the macromolecular nature of coal seems to mean that 'coals are macromolecular gels, i.e. have three-dimensionally cross-linked structures'. Coal contains relatively high concentrations of trace elements/heavy metals when compared with other geological materials (Fernandez-Turiel *et al*., 1994). General proximate analyses shows that the organic component of coal is predominantly composed of carbon which is 70-90% with significant amounts of hydrogen, oxygen, nitrogen and sulfur (usually in the range 1-

10%) and traces of many other elements (Graham and Trotman, 1983). Heavy metals in coal may be classified according to whether they are primarily associated with the organic fraction, with the mineral fraction or with both. Most elements of environmental concern including arsenic (As), cadmium (Cd), mercury (Hg), lead (Pb) and zinc (Zn) are concentrated in the organic fraction of the coal. Some, such as chromium (Cr), copper (Cu) and selenium (Se) were present in both mineral and organic matter, but these also tended to be inorganically associated; boron (B), beryllium (Be) were found to be largely combined with the organic fraction of coal (Chadwick *et al*., 1987).

Goldschmidt (1935, 1937) suggested three main stages for the enrichment of trace elements/heavy metals in coal, namely concentration during plant growth, during decay of vegetable matter and during mineralization. These are in keeping with a classification of mineral matter in coal put forward later by Swaine (1962a, 1975) who reviewed a wide range of publications relevant to the topic. Szadeczky-Kardoss and Vogl (1955) who had studied several coals stated that trace elements enrichment took place during the formation of peat, namely the uptake of elements by plants, the removal of elements by ion exchange from swamp waters, the direct addition of mineral matter from incoming waters and from the atmosphere and minerals *in-situ*. According to Allen and van der Sande (1984), lignite coal is rich in Ba and Fe, bituminous coal is rich in Ti and semi-anthracite coal is rich in Ti and Ca. Bethell (1962) had concluded that trace elements generally decreased with increasing rank of coal whereby the rank of coal increases from lignite coal, subbituminous coal, bituminous coal to anthracite coal (Speight, 1994).

2.3 Mobilization of Trace Element by Coal Utilization

2.3.1 Coal-fired Power Plants

Coal has a significant role in energy production and represents a considerably quantity of electric energy uses coal (Kovacs and Mang, 2002). The annual world coal produced in 1978 was estimated to be 2.974 x 10^9 tonnes and about 4 billions tonnes of black coal and 800 million tonnes of brown coal and lignite are produced in the world annually (Quenton, 1980; Kovacs and Mang, 2002). Coal power plants generate about 23% of the electricity consumed world-wide (EIA, 2006). A single large power plant (>2000 MW) will burn as much as 10^7 tonnes of coal a year (Wiersma and Crockett, 1978). About 110 million tonnes of coal were used in power stations during the year 1989-1990 generating nearly 35 million tonnes of ash (Palit *et al*., 1991). Depending on the ash content of the coals, which varies between 5-8% and 30-35%, the quantity of solid and fly ash produced by firing is 1.0-1.5 billion tonnes per year. The quantity of residues of this kind accumulated in the past amounts to 100 billion tonnes (Kovacs and Mang, 2002). With a yearly increase of nearly 15% in thermal generation, 125 million tonnes of ash could be generated per year by the turn of the century (Palit *et al*., 1991). Although potentially toxic elements are generally present at relatively low concentrations in coal, significant mobilization still occurs because of the very large quantities of coal consumed (Chadwick *et al*., 1987).

Leaching of heavy metals to the surface and underground water source are one of the major concerns about coal combustion residues (CCRs) disposal which may contaminate

the ground water quality nearby the ash disposal area (Anderson *et al*., 1993; Sandhu *et al*., 1992). The trace elements in the coal residues, such as Zn, Cd, Pb, Mo, Ni, As, Se and B are an important concern for land disposal due to their environmental significance (Keefer, 1993; Spears, 2000). However, ultimate impact of each trace element will depend upon its state in coal combustion residues (CCRs) and toxicity, mobility and availability in the ecosystem (Asokan *et al*., 2004). Thus, the production of energy in coal-fired power plants mobilizes an important variety and quantity of trace elements, some of them being potentially toxic to the biological system including Cd, Co, Cu, Ni, Pb, Sb and Zn (Sabbioni and Goetz, 1983).

2.3.2 Behavior of Trace Element within Power Plants

The behavior of trace elements in power plants has involved a detailed examination of the portioning of trace elements between the various exit streams, to allow an assessment of their flow rates (Chadwick *et al*., 1987). Mass-balance studies have been carried out for a whole range of trace elements (Gladney, 1974; Kaakinen *et al*., 1975; Klein *et al*., 1975; Schwitzgebel *et al*., 1975). Studies have shown that coal combustion in power plants produces a consistent and selective partitioning of trace elements between the various exits stream and can be classified into three groups which are Group I, Group II and Group III. Elements in Group I are partitioned equally between bottom ash and are not concentrated in the outlet ash. Elements in this group are Al, Ba, Ca, Ce, Cs, Fe, K, Mg and Th. Elements in Group II are concentrated in the inlet fly ash compared to the bottom fly ash and the outlet fly ash to the inlet fly ash. This group consists of As, Cu, Pb, Se and

Zn. Group 3 are whereby the elements are mainly present in the gas phase and are depleted in all ashes. This group consists of three elements such as Br, Hg and I.

Several elements, Cr, Ni, U and V, exhibit intermediate behavior between Group I and Group II (Chadwick *et al*., 1987). Metals such as Al, Ca and Fe may be found in rather high concentrations, up to and over 1% of the total coal weight but these are largely bound into the ash and remain there after combustion (Graham and Trotman, 1983).

2.4 Coal Ash

2.4.1 What is Coal Ash?

Coal ash can be described as rather inert, almost wholly insoluble solid which is composed mainly of silica, alumina and lime with some proportion of iron oxide together with trace amounts of other minerals, the exact proportions varying according to the coal source (Graham and Trotman, 1983) as shown in Table 1. Trace elements make up the rest of the ash (Davison *et al*., 1974). The composition of the ash which are 50% Si and oxides of Al, Fe and Ca makes up 95-99% of its contents whereas Mg, Na, K, Ti, S and C account for 0.5-3.5% (Mehra *et al*., 1998). Oxides of Al, Ca, Fe and Si are the predominant constituents and the element carbon (soot, carbon black) is a significant fly ash constituent.

The ash released from a coal-fired power station is generally referred to as coal fly ash (CFA). However, this term seems to be confusing due to two reasons. First, the word ‘fly

ash' is a generic term, which can be applied to residues from the combustion of coal depend on the combustion technique used and to a considerable extent upon the precise part of the plant in which the ash is produced (Meij and Te Winkel, 2007). The production of coal ash is one of the major environmental problems associated with the use of coals as fuel in thermal power plants and is a serious source of air pollution since it remains air borne for a long period of time and causes health hazard (Mandal and Sengupta, 2006; T.E.R.I, 1998). To prevent coal ash from being air borne, the dumping ground needs to be kept wet at all times (Gupta *et al.*, 2005). The main method of disposal of fly ash from these power stations is by mixing the ash with water, the resultant slurry is pumped in through pipes to ash disposal ponds (Mehra *et al.*, 1998).

Table 2.1: A summary of the constituents of fly ash (Graham and Trotman, 1983)

Main Constituents of Fly Ash	**Percent Compositttion**
Silica (SiO_2)	43-54%
Alumina (Al_2O_3)	22-32%
Fe_2O_3/TiO_2	4-15%
CaO	1-8%
$MgO/H_2O/Na_2O$	3-9%
SO_3	0.2-2%
Source: Les Cendes Volantes des Centrales Thermiques Electricite de France, Paris,1979	

2.4.2 Types of Coal Ash

Combustion of coal in coal-fired power plants produces two main of solid wastes which are fly ash and bottom ash. Bottom ash is the course-grained fraction that is collected from the bottom of the boiler and is disposed of by the wet disposal method in a slurry form to nearby waste disposal sites (ash ponds) (Mandal and Sengupta, 2006). Fly ash is a fine powdery residue contains Si, Al, oxides of Fe, Ca, Mg, As, Cr, Pb, Zn, Ni and other toxic metals (Mehra *et al*.,1998; Palit *et al*.,1991). It consists of finer sized particles, ranging from 0.5 to 200 μm and is the most important waste materials in modern pulverized coal power plants (Baba, 2002; Chadwick *et al*., 1987). Owing to its relatively small size and, hence, large surface area, the ashes have a greater tendency to absorb trace elements that are transferred from coal to waste products during combustion (Gilec *et al*., 2001).

Depending upon the parent coal and operating conditions of the furnace, fly ash varies in its physical, chemical and mineralogical composition (Adriano *et al*., 2000; Asokan, 2000; McCarthy and Dhar, 1999). Chemical composition of ashes and slags depends on the sort of crude mineral substance existing in hard coal (Baranowska, 2000).The proportion of fly ash, bottom ash and slag that were produced will vary not only with the ash content of coal, but with the plant design and the particulate collection device employed. Ash that was removed in a wet condition may have considerably higher elemental enrichment. The total ash produced in cyclone-fed units are divided equally between bottom ash and fly ash, while in the more commonly occurring pulverized coal plants over 90% of the total ash may be fly ash (Bolton *et al*.,1973). Fly ash particles are

considered to be highly contaminating as their high surface area gives rise to the enrichment of potentially toxic trace elements which condensate during cooling of combustion gases (Klein *et al*., 1975; Coles *et al*., 1979).

2.4.3 Enrichment of Trace Element in Fly Ash

The enrichment factor (EF) is expressed as the extend of trace element enrichment on fly ash (Chadwick *et al*., 1987). This is the ratio of the concentration of an element (X) to that of Al, or another refractory element in the fly ash, divided by the corresponding abundance ratios in crustal rocks (Mason, 1966). The equation of enrichment factor (EF) is as below:

$$EF = [([X] / [A]) \text{ fly ash}] / [([X] / [A]) \text{ crust}]$$

This normalization technique takes into account the apparent enrichment of fly ash caused by carbon losses in the combustion of coal (Chadwick *et al*., 1987). The enrichment of trace elements in the ash depends on the physico-chemical properties of the elements, their chemical form in the coal, processes of coal combustion and mechanisms of emission control devices (Kaakinen *et al*., 1975; Phung *et al*., 1979; Fulekar *et al*., 1983).

2.5 Environmental Effect of Coal Ash

2.5.1 Atmospheric Emission of Trace Element

Trace elements are emitted into the air in solid (fly ash) and gaseous state (Meij and Winkel, 2007). In general terms, it seems likely that emission of trace elements as vapor, rather than absorbed on particulate, is preferable as this would increase atmospheric dispersion. There is reasonably good agreement on the set of elements that are preferentially emitted in the vapor phase or on fly ash. Elements that are largely emitted on fly ash are Cu, Zn, Mo, Cd, Pb, Cr, Na, Ni, Cs and U. Element such as As, Se, Ga are emitted on fly ash and as vapor. Elements which are emitted largely in vapor phase are Hg, Cl and F (Graham and Trotman, 1983).

Heavy metals are released into the surrounding environment during the combustion in power plants are by two different pathways. First pathway is atmospheric emissions via stack, including elements which are volatized and those which are condensed and enriched with free particles that escape from the particulate controls. It is concluded that fly ash dispersal from the stacks are a source of alkali, alkaline earth and to some extent heavy metals in soils in the vicinity of the power stations (Mehra *et al*., 1998). Second is the leaching of combustion wastes (fly ash and slags) once these are ponded or landfilled which result in the enrichment of elements in river overbank soils (Fernandez-Turiel *et al*., 1994; Mehra *et al*., 1998).

In recent years several field investigations have assessed the magnitude of atmospheric trace element deposition from power plants (Chadwick *et al*., 1987). Smith (1980) points out that volatilized elements which condensed upon fly ash (As, Sb, Pb, Cd, V, Mo, Zn, Ga, Cs and U) are often emitted into the atmosphere by a factor of up to 10 or more than elements not volatilized and thus have a higher probability of pulmonary deposition upon respiration.

Particulate emissions from coal combustion may be inhaled by humans and one of the aspects of human health concern is the possible carcinogenic properties of ash (UNGA, 1980). The three elements As, Cd and Ni possess carcinogenic forms of importance as As(III), Cr(VI) and Ni carbonyl and are present in high concentration in fly ash particles (Chadwick *et al*., 1987). There is no information available on the abundance of the carcinogenic forms of As and Cr in fly ash, but Van Hook (1978) considers that both are present.

2.5.2 Solid Wastes

The combustion of coal in power plants produces solid wastes which are fly ash, bottom ash and slag and, if SO_2 scrubbers are employed, scrubber sludge (Chadwick *et al*., 1987). These solid wastes are also known as coal combustion residues (CCRs) (Asokan, 2003; Keefer, 1993). During the combustion, the wastes are enriched from 4 to 10 times for some of the trace elements (Fernandez-Turiel *et al*., 1994). Fly ash generally contains higher metal concentrations, which makes it a potentially greater threat to the

environment compared to bottom ash (Landsberger *et al*., 1991). Therefore, fly ash is the most important waste material in modern pulverized coal power plant and ash retention may vary between 70 and 90% (UNGA, 1980; Chadwick *et al*., 1987). The leachability of fly ash is enhanced for several reasons (Landsberger *et al*., 1991). Studies have shown that most of the elements present in coal ash, e.g. 8% of Ni to 17% of Cr are able to leach (Qeurol *et al*., 1993). The smallest particles have the greatest total surface area, thus providing more sites for the adhesion of potentially toxic trace elements (Landsberger *et al*., 1991). The more volatile elements are preferentially deposited on the surface of the fly ash particles (Ontiveros *et al*., 1989). Such a high percentage of heavy metals in fly ash may reach human beings through plants and can cause serious adverse effects (I.A.R.C., 1980, 1987, 1990).