2015 World of Coal Ash

Characteristic Studies of the Toxic Hazardous Elements in Coal Ash from Coal Power Plants and Comparison of Circulating Fluidized Bed Boiler (CFB) and Pulverized Coal-fired Boiler (PCB) Technologies

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ABSTRACT

After the accident of nuclear power plants in Fukushima, Japan, globally coal power plants has growing demand and number of new-built coal power plants has established. CO2 emissions are increased with the increasing of new coal power plants. The United States, which has 1,100 coal ash sites, produces 140 million tons of coal ash from coal-fired power plants every year. Dumping of coal ash has brought out environmental problem and health hazards because coal ash contains toxic heavy metals. To reduce the landfill of coal ash, many countries have undertaken recycling of coal ash as key resources for recovery valuable critical rare earth elements and utilize the coal ash as construction material. One of our objectives was to evaluate the characteristics of hazardous heavy metals particularly mercury, arsenic, chromium and lead presented in coal combustion products (coal fly ash and bottom ash) from coal-fired power plants. The aim of this research was to identify and compare the technologies between Circulating Fluidized Bed Boiler (CFB) and Pulverized Coal-fired Boiler (PCB). In this study, coal and coal ash samples from different areas (Indonesia, Japan and Korea) are used for the evaluation of heavy metals concentration.

INTRODUCTION

Coal is an important resource that accounts for 29.9% of global primary energy. Coal produces 41% of all electricity around the world and is used in 70% of steel production globally.[1] Since the Fukushima Daiichi nuclear power plant disaster in Japan, the global trend has been shifting away from the construction of new nuclear power plants.[2] Combined with increased electricity demand and advancements in electrical generation technology, the abundant supply of coal reserves and its low cost have led to a rise in coal demand.[3] Looking at total global coal production by year, coal consumption rose from 4677Mt in 1990 to 7608 Mt in 2011, and to 7830Mt in 2012.[4]

Along with this trend, coal ash dumping is becoming a serious problem in countries around the world. The U.S. produces over 92 Mt of coal ash annually, 40% of which is recycled in various applications while the other 60% goes into storage or to disposal sites, leading to environmental issues.[5]

Raw coal used in coal-fired power plants, which largely determine the composition and properties of coal combustion products, contains about 120 different minerals. Among these, several elements including Cd, Pb, and As have the potential to be hazardous.[7] Toxic materials contained in coal ash can differ according to the location where the coal was extracted. Typical materials found in coal ash include arsenic (As), lead (Pb), mercury (Hg), cadmium (Cd), chrome (Cr), selenium (Se), aluminum (Al), antimony (Sb), barium (Ba), beryllium (Be), boron (B), chlorine (Cl), cobalt (Co), manganese (Mn), molybdenum (Mo), nickel (Ni), thallium (Tl), vanadium (V), and zinc (Zn).[8] Such toxic materials can cause damage to major organs in humans and can adversely affect human health and development as well as other living organisms in the ecosystem.[12]

In order to address the above problems, countries around the world are working to develop technologies for environmentally friend coal-fired power generation. Examples of such environmentally friendly and highly efficient coal-fired power generation technologies include ultra supercritical pulverized coal-fired power generation (USC-PC), integrated gasification combined cycle (IGCC), and pressurized fluidized bed combustion (PFBC) (Fig 1).[19]

Highly efficient, environmentally friendly and high capacity 600℃ grade ultra supercritical coal-fired power plants have been in commercial use in developed European countries and Japan since the 1990s, based on research and development going back to the 1980s.[3, 17] Current research in Korea is focused on the development of highly efficient, high capacity and environmentally friendly technologies.[17]

This study is carried out in the context of the growing use of low rank coal alongside the development of environmentally friendly coal-fired power generation technologies in Korea. It provides an overview of the current situation of power generation in Korea. As Korean researchers are working to develop fluidized bed boilers as a next generation technology with high efficiency, this study compares the characteristics of the fluidized bed boiler and the pulverized bed boiler in order to offer a better understanding of the developments in this field. Finally, the behavioral characteristics and concentrations of heavy metals within the waste products of coal-fired power plants are discussed.

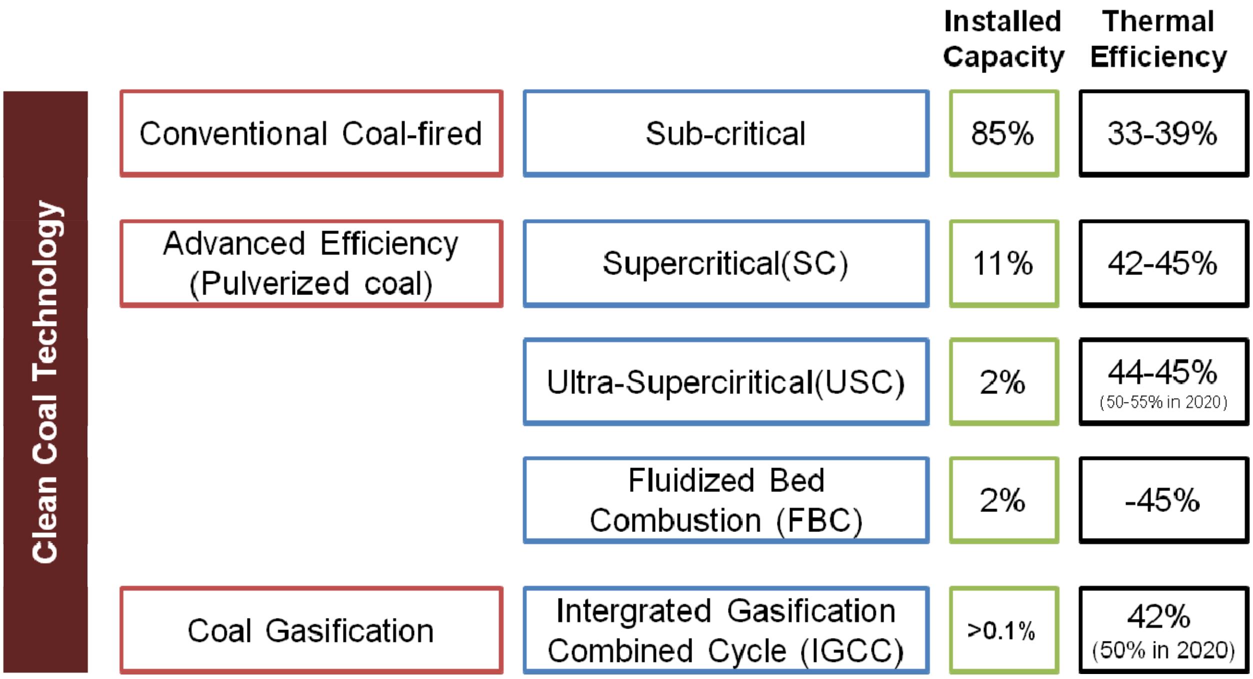

Fig 1. Clean coal technologies for Coal-fired power generation with installed capacity and thermal efficiency. Countries which have advanced power generating technologies focus on developing eco-friendly and efficient coal-fired power generation tehcnologies such as USC, FBC, IGCC, etc.[19] (Energy Technology Perspective 2006, IEA)

1. OVERVIEW OF THE SITUATION OF COAL-FIRED POWER PLANTS AND PRODUCTION OF COAL ASH IN SOUTH KOREA

The rapid growth of key industries such as machinery, electronics, automobiles, steel, and petrochemicals in Korea over the past decade was accompanied by a corresponding growth in the proportion of industrial demand in total consumption. Total electricity consumption grew at an annual rate of 5.6% in the 11 years from 2002.[4] As coal-fired power plants offer advantages in terms of lower investment costs and fuel costs as well as shorter construction periods, more Korean corporations are entering the business of coal-fired power generation (Fig 2). In 2013, the Korean Ministry of Knowledge Economy released its 6th Basic Plan of Long Term Electricity Supply & Demand.[5] According to the plan, by 2027, 15,300MW out of 29,570MW of newly installed capacity will come from coal-fired power plants with a total of 23 new facilities planned for construction (Fig 2).

Since the restructuring of the electricity market in 2001, power companies have been working to combat increased competition by reducing the cost of raw materials, which takes up the largest proportion of costs involved in coal-fired power generation.[14] Following fuel price fluctuations in 2007 and 2008, power companies greatly increased the proportion of low rank coal in fuel imports in order to lower costs and this has since been a continuing trend. Amid rising oil prices in the global energy economy, fluidized bed combustion is generally seen as the most appropriate technology to achieve environmentally friendly energy consumption and highly efficient combustion of the low rank anthracite used in domestic power plants.[13]

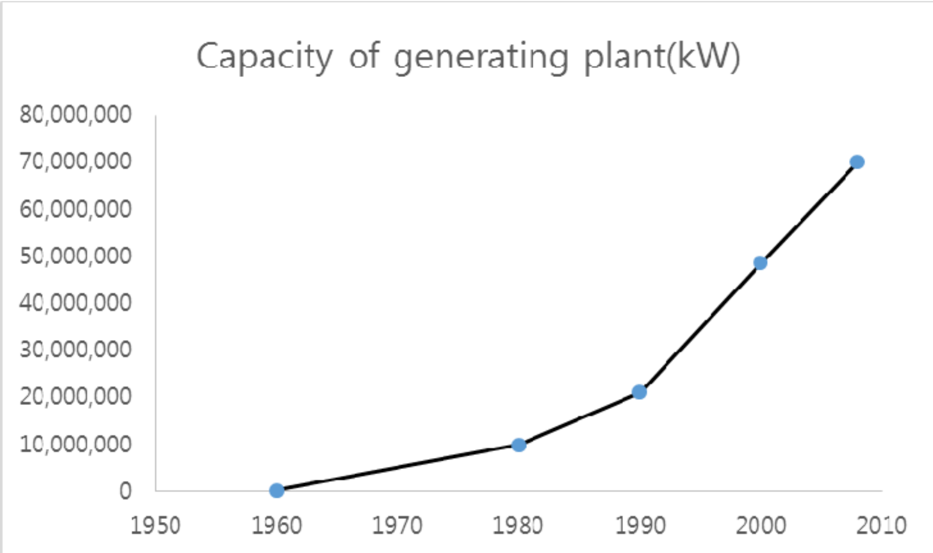

Fig 2. Increased trends of construction of Coal-fired power plants in South Korea [4]

In Korea, coal ash production is increasing due to increasing numbers of coal-fired power plants to meet the rapidly growing electricity demand. The rise was most pronounced in 2011. Although recycling rates are increasing steadily (Fig 3), they remain at a lower rate than other developed nations and areas of application are still limited. Thus, there is a growing trend of transition to circulating fluidized bed boilers, which are environmentally friendly boilers that produce electricity at high efficiency and low cost.

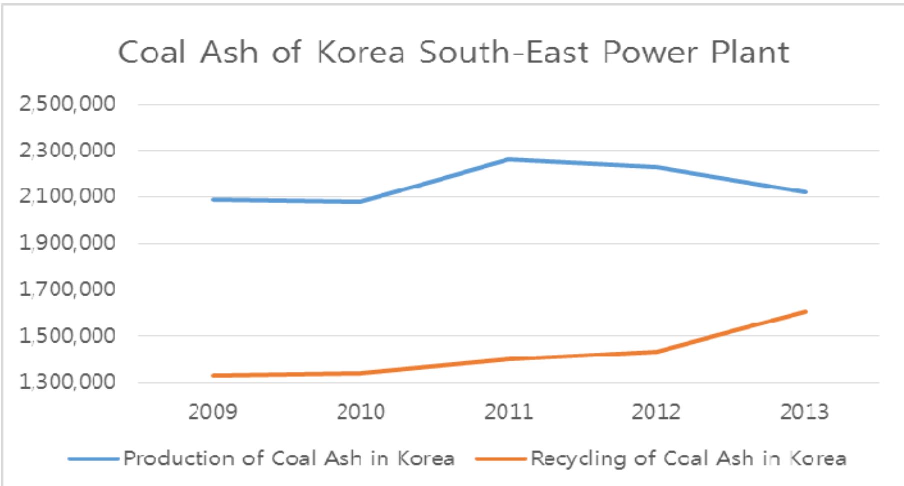

Fig 3. Production(Blue) and Recycling(Red) of Coal combustion product(Coal ash) from South-East Power Plant including Samcheonpo power plant, Yeongheung power plant, Yeongdong power plant and Yeosu power plant located in South-east part of South Korea (unit: ton), Ref: http://www.kosep.co.kr/

2. THE TYPES OF COAL-FIRED POWER PLANTS AND THE CHARACTERISTICS OF COAL COMBUSTION PRODUCTS

2.1. COAL-FIRED POWER PLANTS: COMPOSITION OF PULVERIZED COAL COMBUSTION AND FLUIDIZED BED COMBUSTION

Pulverized coal combustion is the most widely-used technology in coal-fired power generation,[11] mostly found in high capacity thermal power plant boilers. The advantages of pulverized coal combustion boiler are that it operates at atmospheric pressure and materials travel through the plant via simplified routes.[11] However, pulverized coal combustion is inferior to fluidized bed combustion when it comes to efficiency and economy of operation. This is because the combustion constraints of low grade fuel necessitate the use of supplementary fuels (oil) to produce flames.[13]

The unique combustion principle of fluidized bed combustion can better overcome the limitations of coal compared to traditional coal combustion technologies. The technology allows for a wider range of fossil fuels to be used with higher efficiency in power generation, thus reducing fuel consumption.[13]

In fluidized bed combustion, jets of air are blown through the bottom of a layer made of inactive particulate materials such as lime, coal ash, and sand. This leads to the formation of a fluidized bed of particulate materials moving up and down. Flammable materials such as coal are combusted within this fluidized bed. Common types of fluidized bed boilers include the bubbling fluidized bed boiler, the circulating fluidized bed boiler, and the pressurized fluidized bed boiler. The speed of fluidization increases in the order of bubbling fluidized bed → circulating fluidized bed → air stream transfer.[12] The circulating fluidized bed boiler is composed of a circulating fluidized bed combustion chamber, a high temperature ash separation device (cyclone or impact separator, etc), a device to return particles back to base (loop seal, L-valve, etc), and depending on the boiler type, an external fluidized bed heat exchanger and fuel feeder (Fig 4).

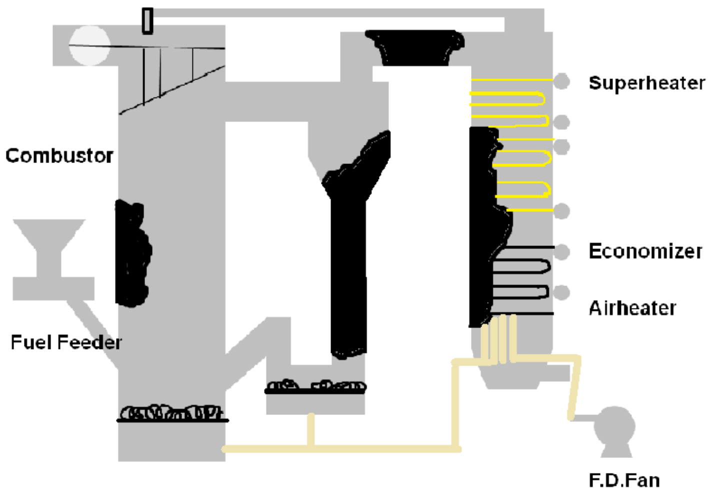

Fig 4. Conceptual Diagram of Fluidized Bed Boiler, Ref: Korea Institute of Energy Research

The main advantages of the fluidized bed boiler are listed in the table below.

- As fuel is surrounded by inactive materials (coal ash, limestone, sand, etc), combustion is not affected by changes in fuel type, water content, and ash content. Thus lower rank fuels unsuitable for existing combustion boilers can be used.
- Temperatures within the combustion chamber remain relatively low at 750~950℃ compared to existing combustion boilers, thus producing lower NOx emissions.
- The fluidization of solid particles minimizes obstruction from slagging, allowing the heating surface to remain clean and eliminating localized thermal stress.
- When using high sulfur content fuels, limestone can be introduced in the combustion process to eliminate 90% of SOx emissions.

2.2. THE PRODUCTION PROCESS AND THE CHARACTERISTICS OF COMBUSTION PRODUCTS FROM COAL-FIRED POWER PLANTS

The process of coal combustion in coal-fired power plants can be divided into three stages (Fig 5). The first step consists of applying heat to coal and the emission of volatile materials. The second and third steps consist of burning the emitted volatile materials and the remaining char.[10] Thus, coal combustion products such as fly ash, bottom ash, boiler slag, fluidized bed combustion (FBC) ash, or flue gas desulphurisation (FGD) material are produced from coal-fired power plants.[9]

Major minerals in coal consist of four mineral groups – aluminosilicates (mainly Kaolinite), oxides (quartz SiO_2 and hematite Fe_2O_3), carbonates (calcite $CaCO_3$, syderyt $FeCO_3$, and dolomite $CaCO_3MgCO_3$), and sulphur compounds (pyrite FeS_2, gypsum $CaSO_42HeO$). Major elements in coal combustion products are SiO_2, Al_2O_3, Fe_2O_3, and CaO [9], which correspond to the mineral composition of coal. Thus it can be seen that the composition and properties of coal ash originate from raw coal.[16]

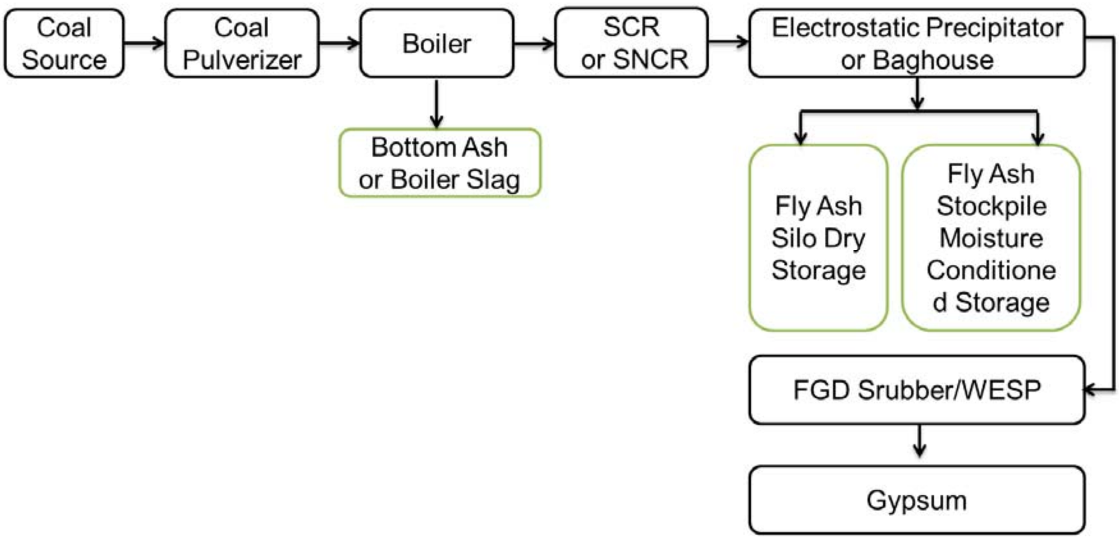

Fig 5. General Ash Generation Process in a Coal fired power plant

3. THE OCCURRENCE AND BEHAVIOR CHARACTERISTICS OF HAZARDOUS METALS IN COAL ASH

There are five major parameters that influence collected ash composition in coal-fired power plant waste products. Ruud Meij and Henk te Winkel (2007) identified the five primary parameters and secondary parameters of influencing factors.[17] For the first primary parameter of fuel, the secondary parameters are type and origin of coal, ash content of coal, the stages of peat, industrial and domestic waste, etc. The secondary parameters of the combustion technique are grate boilers, pulverized coal boilers with dry ash removal, pulverized coal boilers with wet ash removal, cyclone boilers, gasification, fluidized bed combustion, fluidized bed gasification, etc. The third primary parameter is temperature and period of residence, with the secondary parameters being during combustion and in the flue gas. The fourth is the type of particle filter, with the secondary parameters of ESPs (high-temperature, low temperature, dry and wet systems), cyclones, baghouses, wet systems, and filter efficiency. The last primary parameter is other flue gas clean-up systems with secondary parameters of deNOx, carbon injection, etc. Among the above parameters, the parameters that have the greater impact on the heavy metal concentration in coal-fired power plant products is the type and origin of coal and the stage of peat, while the other parameters have supplementary effects. Thus, there is a direct relationship between the concentration of heavy metals in atmospheric power plant ash emissions and heavy metal content in raw coal. [13]

With the passing of the Clean Air Act of 1990, the U.S. Environmental Protection Agency (USEPA) conducted a study on 15 elements out of the 120 elements of coal (Be, K, Cr, Mn, Co, Ni, As, Se, Cl, Cd, Sb, Hg, Pb, Th, U) as potential Hazardous Air Pollutants (HAPs), drawing the conclusion that out of the 15, Hg and As are causes of human health problems.[7] Trace elements within coal are emitted during the combustion process in bottom ash, fly ash, and flue gas.[16]

Most heavy metals have high boiling points and thus are emitted as solid particles in bottom ash or fly ash, being easily eliminated in air pollution prevention facilities for particulate materials.[15] However, mercury has high volatility and a low boiling point, and thus exists in a gas form within gaseous emissions. This makes it difficult to eliminate in facilities to control particulate emissions, allowing it to pass through the filters into the atmosphere.[13]

Trace elements bound within coal are classified into Class 1, Class 2, and Class 3 depending on their distribution behavior.[16,18] Class 1 elements include Cd, Mn Pb, and Fe, which do not evaporate during combustion and remain incorporated in the bottom ash.[16,18] Class 2 elements, which are more concentrated in fly ash than bottom ash, include As, Se, Co, Cr, Cu, and Zn.[16,18] Class 3 elements are highly volatile elements such as Hg that remain in gas form. [16,18]

4. EXPERIMENTAL RESULTS

4.1. MATERIALS AND METHOD

In order to study the heavy metal content and behavior of coal and coal ash, samples of coal (Hadong Coal), bottom ash, and fly ash from a fluidized boiler (Yeosu ash-B/A and Yeosu ash-F/A), and bottom ash from a pulverized coal boiler (Taean ash-B/A) were analyzed. Hadong coal, provided by Hadong Thermal Power Site Division of Korea Southern Power Co., Ltd., was imported from Long Daliq, Long Iram, West Kutai, East Kalimantan. Yeosu ash-B/A and Yeosu ash-F/A were provided by the Yeosu Thermal Power Site Division of Korea South-East Power Co., Ltd., which uses a fluidized bed boiler system. Taean as-B/A was supplied by the Taean Thermal Power Complex Division of Korea Western Power Co., Ltd.

An XRF analysis was conducted to determine the chemical content of each sample. The XRF analysis was commissioned to the Geoanalysis Center of the Korea Institute of Geoscience and Mineral Resources, R&D Tech-Biz Division. Test conditions were set at 24 ± 3 ℃ temperature, and 25 ± 5% R.H. humidity. The heavy metal content analysis was commissioned to the Center for Chemical Analysis at the Korea Research Institute of Chemical Technology and conducted using the ICP-AES test method.

4.2. RESULTS AND DISCUSSION

4.2.1. MAJOR AND MINOR COMPONENT OF COAL AND COAL ASH

Coal contains about 120 types of different elements, of which 33 are common, and 8 types make up the majority.[7] Results of the XRF content analysis to identify the basic components of coal are compiled in the table below.

Table 1. Major and minor chemical composition of samples by XRF (unit: wt.%)

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	MnO	P_2O_5	Igloss
Hadong Coal	2.4	16.85	47.93	1067	2.44	0.11	0.23	0.6	0.77	0.04	12.42
Yeosu ash-B/A	42.51	4.55	8.51	29.62	2.84	0.4	0.72	0.22	0.11	0.1	1.85
Yeosu ash-F/A	35.05	14.04	10.52	25.65	5	0.86	0.35	0.9	0.15	0.11	3
Taean ash-B/A	57.84	20.77	9.11	4.29	1.32	1.08	0.92	1.13	0.1	0.31	3.44

The major mineral components of coal consist of quartz (SiO_2), aluminosilicates, calcite, siderite with iron content, and pyrite with sulfur content.[19] The XRF analysis showed that Hadong coal had higher Fe_2O_3, unlike ordinary coal which has high Si and Al content. This appears to be due to the orogeny of the Central Kalimantan Range and operations of Muller Mts in Kalimantan, Indonesia.[19] Yeosu ash B/A, Yeosu ash-F/A and Taean ash-B/A are products of coal combustion in the respective power plants. Yeosu ash B/A and Yeosu ash-F/A from a fluidized bed boiler had CaO values of 29.62 wt.% and 25.65 wt.%, respectively, while Taean ash-B/A from the PC boiler had a CaO value of 4.29 wt.%, showing a much lower value than that from the fluidized bed boiler.

4.2.1. HAZARDOUS COMPONENTS OF COAL AND COAL ASH

In this study, we investigated the behavioral characteristics of Cr, As, Cd, Pb, Hg. The 5 elements are representative of 15 elements as Hazardous Air Pollutants(HAPs, specified by USEPA) which cause health problems (Fig 6). Hadong Coal and all coal ash samples had less than 0.1 ppm Cadmium(Cd) that can lead to itai-itai disease.

As Lead(Pb) is classified as Class I that remains incorporated in the bottom ash during combustion, lead is accumulated in respiratory organs or bone with adsorption on fine ash. Bottom ash(Yeosu ash-B/A) and fly ash(Yeosu ash-F/A), produced from Yeosu Thermal Power Plant, contained 25.3 ppm and 1.3 ppm of Pb relatively. From that result, it is clear that lead is more concentrated on coarser particles than finer one. Among all samples, Hadong Coal had the highest concentration of lead with 41.2 ppm.

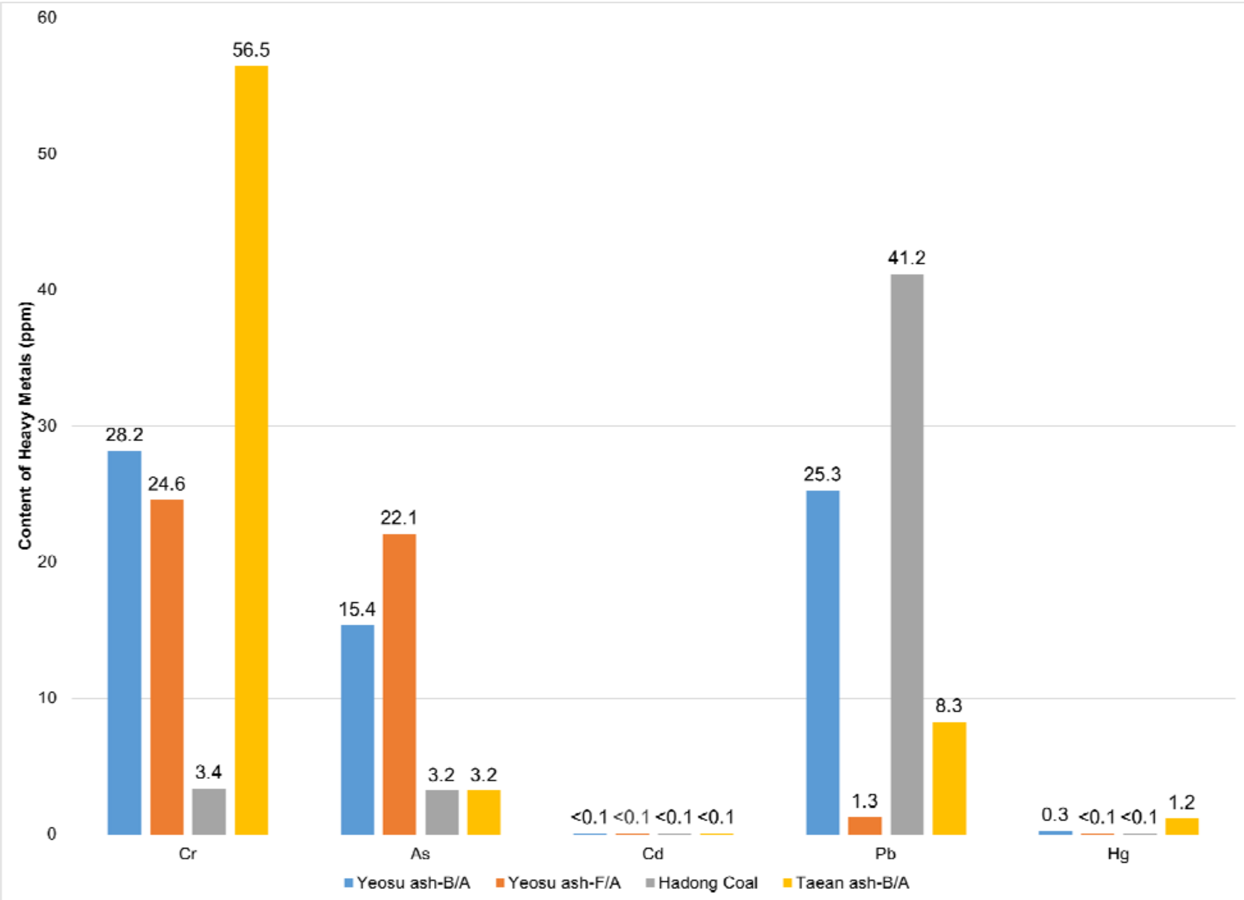

Fig 6. Concentration of Heavy Metal Elements (Cr, As, Cd, Pb, Hg) by ICP-AES (unit: ppm)

Arsenic(As) which is one of Class 2 elements is generally involved as a sulfide in minerals and tends to be more concentrated on the surface of fly ash than that of bottom ash. In Yeosu ash-F/A, 22.1 ppm of As was measured which was 6.7 ppm more As than Yeosu ash-B/A.

Chromium is widely known as carcinogenic, toxic and oxidative substance. Although it is one of Class 2 elements, the samples used in this study showed different behavior comparing to the behavioral feature of Class 2 elements. Particularly, Taean ash-B/A produced from Taean Thermal Power Plant contained very high concentration of Cr with 56.5 ppm. Hadong Coal had lowest Cr measured as 3.4 ppm among the samples.

Finally, highly volatile mercury(Hg) is classified as a Class 3 element and was measured as very low concentration in all samples used in this study. It is considered Hg volatilized remaining in the gas form during coal combustion in the boiler.

5. CONCLUSION

Globally, coal combustion waste produced from coal-fired power plants causes serious problems on the environment and human due to toxic materials. Countries all over the world endeavor to find the solution by recycling the waste or developing technologies for eco-friendly coal-fired power generation including USC-PC, IGCC, and PFBC. In particular, fluidized bed boiler has shed new light recently with several advantages from the viewpoint of the possibility to use lower rank fuels, operating at low temperature, economic and environmental feasibilities.

With overviewing the current situation of power generation in South Korea, this study identified and compared two major boiler technologies. Also, the behavioral characteristics and concentrations of heavy metals within the waste products of coal-fired power plants by examining the coal and coal ash samples from Korean power plants.

Announcement of the Korean Ministry of Knowledge Economy in South Korea about Electricity Supply & Demand included the plan to install 23 new coal-fired power generation facilities. Since 1960s, the capacity of generation plants is increasing with the growth of industrial fields (electronics, automobiles, etc.). South Korea also has developed environmentally friendly combustion boiler system as the advanced countries. For these reasons, combustion waste production from coal-fired power plants is increasing and their recycled rate remain lower that other developed countries.

In this research, we investigated chemical composition and the behavioral characteristics of hazardous toxic heavy metals in coal and coal ash samples from different coal-fired power plants in South Korea. The major component of ash samples is SiO_2, but Hadong coal has higher amount of Fe_2O_3. Since the type and origin of coal and the stage of peat affects the composition of coal combustion ash, the major, minor chemical composition and trace heavy metals in the ash samples were influenced by the original coal used in the power generation sites. From ICP-AES test method, Taean ash-B/A generated from the Taean Thermal Power Complex Division of Korea Western Power Co., Ltd. contained the highest concentration of Chromium with 56.5ppm. Yeosu ash-F/A from the Yeosu Thermal Power Site Division of Korea South-East Power Co., Ltd. consisted of relative higher concentration Arsenic than Yeosu ash-B/A which meant that chromium behaved as a Class 2 element. Very low value of Cadmium and Mercury were shown. The result showed the heavy metal vaporization and concentration in burning coal are roughly predicted by the general law (Class 1, 2, 3). For the future work, the studies of behavioral properties with size distribution, the mechanism and what compositions are affected by each boiler system are required.

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