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Characteristic studies of coal power plants ash sample and monitoring of PM 2.5

Thriveni. T[1], Ramakrishna. CH[2], Seong Young Nam[3], Chunsik kim[4] and Ahn Ji Whan[5*]

[1,2]Hanil Cement, 302 Maepo-gil, Maepo-eup, Danyang-gun, Chungcheongbuk-do, Korea.

[3,4]Technical Center, HANIL CEMENT Co.Ltd., 50, Daehwa-ro 52beonan-gil, Daedeok-gu, Daejeon, 34361, Republic of Korea.

[5]Mineral Processing Division, Korea Institute of Geoscience and Mineral Resources (KIGAM), Gwahagno-124, Yuseongu-gu, Daejeon, Korea.

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Abstract

Coal power plants produce electricity for the nation's power grid, but they also produce more hazardous air emissions than any other industrial pollution sources. The quantity is staggering, over 386,000 tons of 84 separate hazardous air pollutants spew from over 400 plants in 46 states. In South Korea also, annual coal ash generation from coal- fired power plants were about 6 million tons in 2015. Pollutants containing particulate matter 10, 2.5 (PM10, PM2.5), heavy metals and dioxins from coal-fired power plant. The emissions threaten the health of people who live near these power plants, as well as those who live hundreds of miles away. These pollutants that have long-term impacts on the environment because they accumulate in soil, water and animals. The present study is to investigate the physical and chemical characteristics of coal-fired power plant fly ash and bottom ash contains particulate matter, whose particulate sizes are lower than PM_{10} and $PM_{2.5}$ and heavy metals. There are wide commercial technologies were available for monitoring the PM 2.5 and ultra-fine particles, among those carbonation technology is a good tool for stabilizing the alkaline waste materials. We collected the coal ash samples from different coal power plants and the chemical composition of coal fly ash was characterized by XRF. In the present laboratory research approach reveals that potential application of carbonation technology for particulate matter $PM_{10,}$ $PM_{2.5}$ and stabilization of heavy metals. The significance of this emerging carbonation technology was improving the chemical and physical properties of fly ash and bottom ash samples can facilitate wide re use in construction applications.

Key words : coal ash, particulate matter, heavy metals, carbonation

1. INTRODUCTION

Currently, there are several environmental issues resulting from human impacts on the quality of our environment particularly in 2015 ~2016 such as Climate change issues (global warming, greenhouse gas effect, ozone depletion), Aquatic environmental issues (water pollution, water shortage, ocean dead zones, water diversion, overfishing), Air quality issues (acid rain, air pollution, nuclear pollution [1].

Particulate matter (PM) is a complex pollutant and it contains a mixture of both organic and inorganic particles, such as dust, pollen, soot, smoke and liquid droplets found in the air which are toxic and hazardous. PM air pollution is often discussed in terms of particle size because of the distinct characteristics (origin, chemical species and atmospheric behavior) associated with different particle size classes. Particulate matters widely dived into 3 cate-

†To whom corresponding should be addressed.
Tel : +82-43-750-1399 E-mail : skkang@kgs.or.kr

Table-1: Selected countries emission standards for NOX, SOX and PM from coal power plants

Country	Time period	NOx (μg/m^3)		SOx (μg/m^3)		PM (μg/m^3)	
		Existing	New	Existing	New	Existing	New
Australia	--	--	800	--	200	--	80
China	Hourly	100	50	200/50	35	30/20	10
Germany	Daily	200	150	200	150	20	10
India	--	600/300	100	600/200	100	100/50	30
Indonesia	--	850	750	750	750	150	100
Japan	--	410	200	--	200	100	50
South Africa	Continuously	1100	750	3500	500	100	50
Thailand	--	820	410	2002	515	180	80
USA	Daily	135	95.3	185	136	18.5	12.3
EU IED	Continuously	200	150	200	150	20	10

gories such as coarse particles in the range of about 2.5 to 10 μm in diameter - combustion of fossil fuels such as coal, oil, near roads mineral dust and petrol can produce. Fine particles (excluding ultrafine particles) in the range of about 0.1 to 2.5 μm in diameter, sometimes called accumulation mode particles; and ultrafine particles less then approximately 0.1 μm in diameter, sometimes called nuclei model particles.

In many developing countries, greenhouse gases (GHGs), reactive trace gases, particulate matter and toxic compounds emissions from waste combustors are more challenging to national and global inventories. Air pollution is a major emerging global environmental problem. Particulate matter pollution in air affects peoples living health quality tremendously, and it poses a serious health threat to the public health as well as influencing visibility, direct and indirect radiative forcing, climate, and eco systems [2].

Coal power plants are the major sources of pollutant emissions. Over 386,000 tons of 84 separate hazardous air pollutants released from over 400 coal power plants in 46 states of USA [3]. Currently the coal industries faces stringent emission regulations to limits the release of SO_2, NOx, toxic volatile organic compounds, heavy metals, and particulate matter (PM) etc. Particle matter contain any or all of the aforementioned chemical species or their compounds, plus water and biogenic organic species. PM2.5 (particles less than 2.5 μm in diameter), cause not only air pollution but also human respiratory and heart disease and cancer. PM2.5 is the major component of smog in China. Control of PM2.5 emissions and their precursors from coal-fired power plant is necessary to mitigate the environmental and health impacts, especially in countries for power generation coal is the main energy source, such as India, China and South Africa. Most countries regulate emissions based on the plant's age. But the definitions for new plant and existing plants are different [4]. There are several reports published for the source of particulate matter emissions from industrial plants [5-10].

1.1. Environmental and Public Health Effects of PM Emissions

Presently, it is not known which particles have the most significant impact on human health, although theories related to particle size and composition. Suspended particles may have diameters varying from several centimeters (e.g. dust particles), to 0.1 mm. The $PM_{2.5}$ (particle sizes are less than 2.5 μm in diameter) size fraction is considered as one of the possible principal causes of cardiovascular and respi-

ratory illness [11-14].

In Fig.1, the represented areas where particulate material from incomplete combustion processes is deposited in the body. These particles are deposited into the airways in the head region when inhaled. The small fine particles are deposited into lung airways or the tracheobronchial region [15]. If the particulate matters emissioned along with heavy metals

Table-2: Health risks from various pollutants, emission limit values (Suggested by WHO guide lines) for coal power plants (adopted from ref 17 and 18)

Pollutant	Health Risks	Emission limit values
Carbon dioxide (CO_2)	Indirect health impacts from climate change	
Sulphur dioxide (SO_2)	Can affect respiratory system and lung functions, aggravation of asthma and chronic bronchitis, makes people more prone to infections of the respiratory tract; irritation of eyes; cardiac disease aggravated ; ischemic stroke risk	20 μ g/m3 (day) 500 μ g/m3 (10min) Directive 2001/80/EC: 400 mg/m3 (old plants), 200 mg/m3 (new plants)
Nitrous oxides (NOx);	Asthma development (suspected), asthma exacerbation, chronic obstructive pulmonary disease, stunted lung development; cardiac arrhythmias, ischemic stroke. Reacts with VOCs in sunlight to form ground- level ozone.	NO2: 40 μ g/m3 (year), NO2: 200 μ g/m3 (1h) Directive 2001/80/EC: NOx: 500 mg/m3 (old plants) NOx: 200 mg/m3 (new plants)
Particulate matter: coarse particulates (PM10), fine particulates (PM2.5)	Exposure to such particles can affect both lungs and heart, especially fine particles - containing microscopic solids or liquid droplets that are so small that they can get deep into the lungs and cause serious health problems. Numerous scientific studies have linked particle pollution exposure to a variety of problems, including premature death in people with heart or lung disease, nonfatal heart attacks, irregular heartbeat, aggravated asthma, decreased lung function, and increased respiratory symptoms, such as irritation of the airways, coughing or difficulty breathing. Respiratory: asthma development (suspected), asthma exacerbation, chronic obstructive pulmonary disease, stunted lung development (PM2.5), lung cancer; Cardiovascular: cardiac arrhythmias, acute myocardial infarction, congestive heart failure (PM2.5). Nervous system: ischemic stroke.	PM2.5 10 μ g/m3 (year), PM10 20 μ g/m3 (year) Directive 2001/80/EC: (monthly, total dust) 50 mg/m3 (old plants), 30 mg/m3 (new plants) Directive 2008/50/EC: 25 μ g/m3 target PM2.5 (year), 50 μ g/m3 (day) limit PM10, not to exceed on >35 days
Antimony (Sb), Arsenic (As), Beryllium (Be), Cadmium (Cd), Chromium (Cr), Nickel (Ni), Selenium (Se), Manganese (Mn).	Carcinogens (lung, bladder, kidney, skin cancers); may adversely affect nervous, cardiovascular, dermal, respiratory and immune systems. The International Agency for Research on Cancer classifies arsenic and its compounds as group 1 carcinogens.	As: no safe level established; Cd 5 ng/m^3 air; Directive 2004/107/EC: As 6ng/m^3; Cd 5ng/m3; Ni 20ng/m^3 (ambient air)
Lead (Pb)	Damages nervous system of children; may adversely affect learning, memory and behaviour; may damage kidneys, cause cardiovascular disease, anemia.	0.5 μ g/m3 (air) Directive 2008/50/EC: 0,5 μ g/m3 (ambient air)
Dioxins and furans (e.g.,2,3,7,8-tetrachlorodibenzo-dioxin , short TCDD)	Probable carcinogen (stomach cancer); affect reproductive, endocrine and immune systems. Dioxins accumulate in the food chain.	WHO AQ Guidelines value: TCDD 70 pg /kg weight / month tolerable intake (provisional)

it gives high potential risk to human health [16, 17].

The severe health effects from heavy metals associated with air pollutants presented in the Table. 2 [18]. In this table we reported the various health risks from various pollutants, and heavy metals. European Union-wide impacts amount to more than 18,200 premature deaths, about 8,500 new cases of chronic bronchitis, and over 4 million lost working days each year [19].

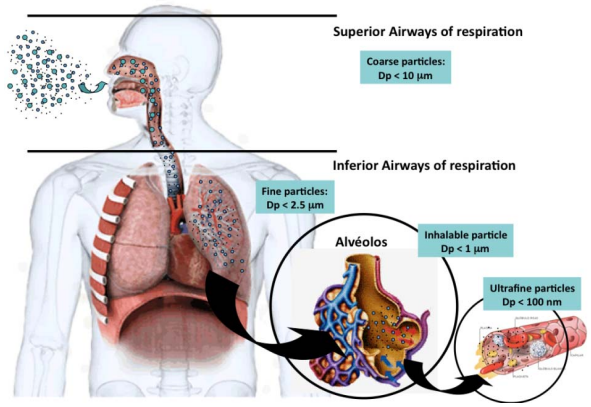

Figure. 1. Health Impacts of Particulate Emissions [adopted from ref.15].

Hazardous emissions threaten health locally and at great distances. Many metals, dioxins and other pollutants adhere themselves to the fine particles. They may travel with airborne particles to distant locations [19].

1.2. Standard methods for sampling and measurement:

The major methods are focusing on fine PM testing and measuring methods concentrate on determin-

Figure. 2. Spatial range of pollutants impact (Adopted from reference 19).

Table 3: Summary of available technologies (Adopted from reference 19).

Technology Name	Pollutants name	Technology working	% of currently using this technology
Wet or Dry Flue Gas Desulfurization (Scrubbers)	HAPs: HCl, HF, HCN, SO2, PM	Liquid mixed with limestone is sprayed into the emission or emissions are passed through a stream of liquid mixed with lime or a bed of basic material such as limestone; reactions between sulfur and base compounds produce salts which are removed from the exhaust air stream.	46%
Electrostatic Precipitators (ESP)	Antimony, Be, Cd, Co, Pb, Mn, Ni, and primary particulate matter	Particles are charged with electricity and collected on oppositely charged plates, particles are collected for disposal/further treatment.	74%
Baghouse	Antimony, Be, Cd, Co, Pb, Mn, Ni, and primary particulate matter	Emissions are passed through fabric filters and collected.	35%
Cyclones	Antimony, Be, Cd, Co, Pb, Mn, Ni, and primary particulate matter	Use centrifugal force to separate particulate from gas streams.	5%

ing the total mass of $PM_{2.5}$. High quality and comprehensive measurement methods for the determination of the chemical components of $PM_{2.5}$ still need to be developed. Since the chemical composition of $PM_{2.5}$ is not completely understood yet, the full chemical analysis of $PM_{2.5}$ remains a challenge.

Currently, there are several technologies are available for the reduction of emissions of particulate matter and heavy metals from coal power plants [19]. The brief summary of available technologies for reductions of emissions from coal power plants presented in Table 3.

In this paper, we presented our investigated research results. We used coal power plants bottom ash and coal ash samples for measuring the particulate matter and

2. EXPERIMENTAL PROCEDURE:

2.1. MATERIALS AND SAMPLE PREPARAITON

In this study, we chosen samcheok green coal power generation CFBC fly ash and bottom ash samples. After receiving the samples we dried the samples at room temperature for the removal of moisture from that samples. After drying, we sieved the coal bottom ash sample by using various mesh sizes of sieving machines. The flow sheet of procedure which we followed as per particle size distribution showed in Fig. 3a, and sieved machine and process as shown in Fig. 3b and 3c.

The coal fly ash and bottom ash samples containing the high content of hazardous heavy metals and stabilized by the carbonation process. In these carbo-

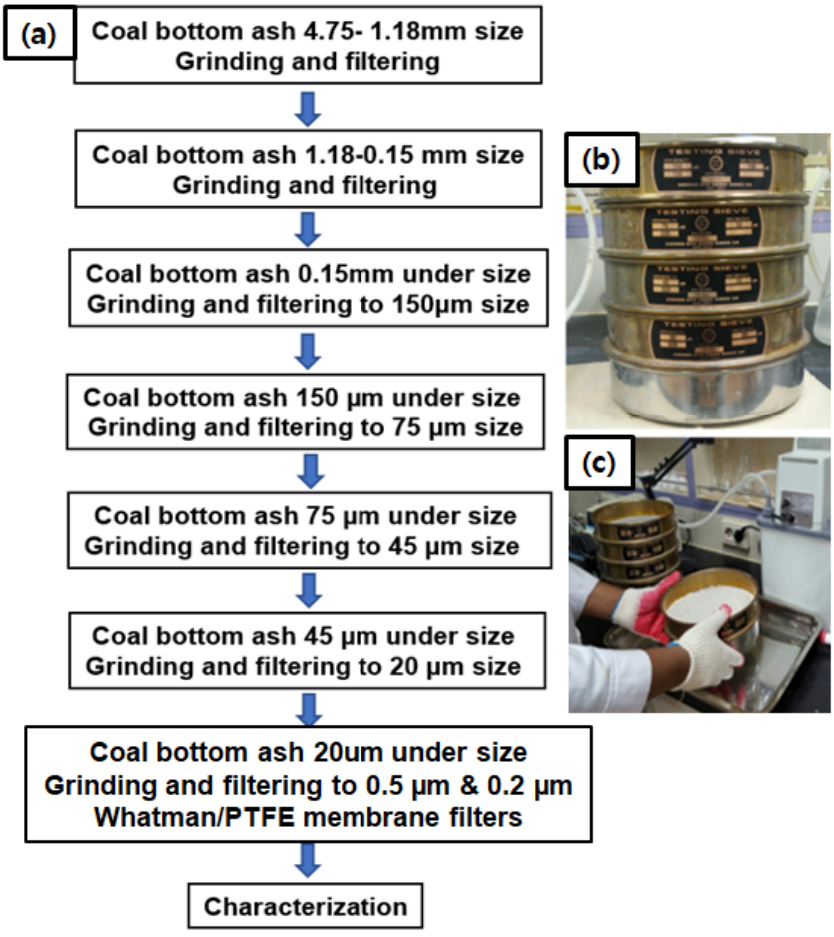

Figure 3. Experimental flow sheet for coal bottom ash sieving (a), sieved machine (b) and process (c).

nation reactions experiments were conducted using a various condition with a liquid-solid-ratio (0.3 and 1.0 dm^3/kg), CO_2 was injected at a rate of 1L/min and 400rpm stirring speed at 20°C temperature. The experiment of carbonation was terminated when the measured pH was below 7 and sample was filtered and dried at 80°C for 12h duration time.

2.2. Carbonation Process:

The aqueous carbonation of calcium hydroxide in contact with compressed CO_2 at moderate temperature allows the synthesis of fine particles of calcite. Carbonation is a strong exothermic reaction. The reaction mechanism of calcite precipitation via aqueous carbonation of $Ca(OH)_2$ was then described by the global reaction.

water

$$Ca(OH)_{2(s)} \rightarrow Ca^{2+} + 2\ OH^- \quad (1)$$

The dissociation of aqueous CO_2,

$$CO_{2(aq)} + H_2O \rightarrow CO_3^{2-} + 2H^+ \quad (2)$$

These processes produce a fast supersaturating (S_I) of solution with respect to calcite,

$$S_I = \frac{(Ca^{2+})(CO_3^{2-})}{K_{sp}} > 1 \quad (3)$$

where (Ca^{2+}) and (CO_3^{2-}) are the activities of calcium and carbonate ions in the solution, respectively, and K_{sp} is the thermodynamic solubility product of calcite.

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3(nuclei) \quad (4)$$

Finally, the crystal growth occurs spontaneously until the equilibrium calcite and the solution is reached

$$CaCO_3(nuclei) \rightarrow CaCO_3(calcite) \quad (5)$$

3. RESULTS AND DISCUSSION:

3.1. Monitoring of particle size distribution in CFBC coal bottom ash sample.

We investigated the physical and chemical characteristics of Samchuk Green Power Circulating Fluidized Bed Power Plant coal bottom ash samples. The CFBC bottom ash contains high amount of calcium because of the desulfurization process using limestone. The bottom ash sample was used to sieving process and the different particle size distribution

Table 4. Particle size distribution ratio coal ash samples

Sieve	1st time		2nd time		3rd time		Average
16mm over	199	12.04%	161.4	9.42%	167.8	10.22%	10.56
16 ~9.50	199	12.04%	238.3	13.91%	177.4	10.80%	12.25
9.50~4.75	178.2	10.78%	223.3	13.03%	226.4	13.79%	12.53
4.75 ~2.36	191.9	11.61%	207.2	12.09%	211.0	12.85%	12.18
2.36 ~1.18	189.6	11.47%	211.7	12.35%	219.4	13.36%	12.39
1.18 ~0.6	132.4	8.01%	150.7	8.79%	162.6	9.90%	8.90
0.6 ~ 0.3	99.9	6.04%	110.8	6.47%	122.0	7.43%	6.65
0.3 ~ 0.15	115.5	6.99%	99.6	5.81%	92.2	5.62%	6.14
0.15 under	347.2	21.01%	310.5	18.12%	263.1	16.02%	18.38
Total (%)	1652.7	99.99%	1713.5	99.99%	1641.9	99.99%	99.98
Total sample weight	1667.2	-14.5	1728.6	-15.1	1654.3	-12.4	

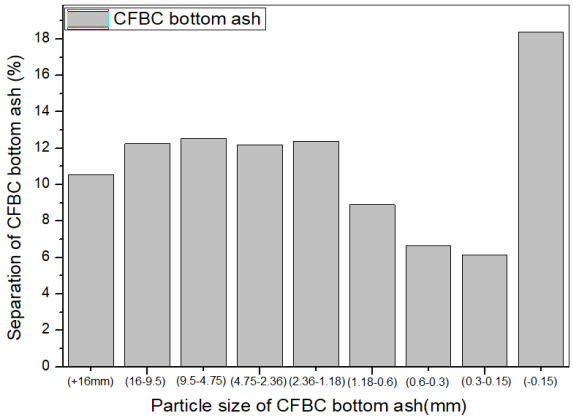

Figure. 4. Particle size distribution of CFBC bottom ash

was shown in Table. 4. These results indicated that bottom ash samples having different particle sizes, among these less than 0.15mm sized particles have high percentage (18.38%) as shown in fig. 4 particle size distribution of CFBC bottom ash sample, these high amount of particle matter having the most significant impact on human health.

3.2. Characteristics of CFBC coal fly ash and bottom ash samples.

X-ray fluorescence (XRF) analysis of the chemical compositions of CFBC coal fly ash and bottom ash samples were presented. Table 5, shows the chemical compositions of CFBC coal fly ash and bottom ash samples. The major components of coal fly ash are 33.3% of CaO and 26.40% of SiO_2 are present and in the case of bottom ash samples having 35.31% of CaO and 39.46% of SiO_2 , remaining elements such as Al_2O_3, Fe_2O_3 and MgO having below 20% are present in both fly ash and bottom ash samples.

The circulated fluidized bed combustion fly ash and bottom ash samples were characterized by XRD and showed in Figure 5. The fly ash collected (differential) through boiler internal dust collector, the residual bottom ash collected from the bottom of boiler (various size). The average amount of 33.3% and 35.31% of CaO is contained in fly ash and bottom ash, respectively. The XRD results shows in fly ash, Lime 20.5%, Periclase 12.3%, Anhydrite 6.4%, Calcite 16.6% and in bottom ash having Lime 9.9%,

Table 5. The chemical compositions of CFBC coal fly ash and bottom ash samples.

Sample name	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	Etc	Ig-loss
CFBC fly ash	26.40	13.40	17.20	33.30	10.00	0.02	0.87	6.57	5.06
CFBC bottom ash	39.46	9.46	6.20	35.31	11.25	0.06	1.25	4.80	4.00

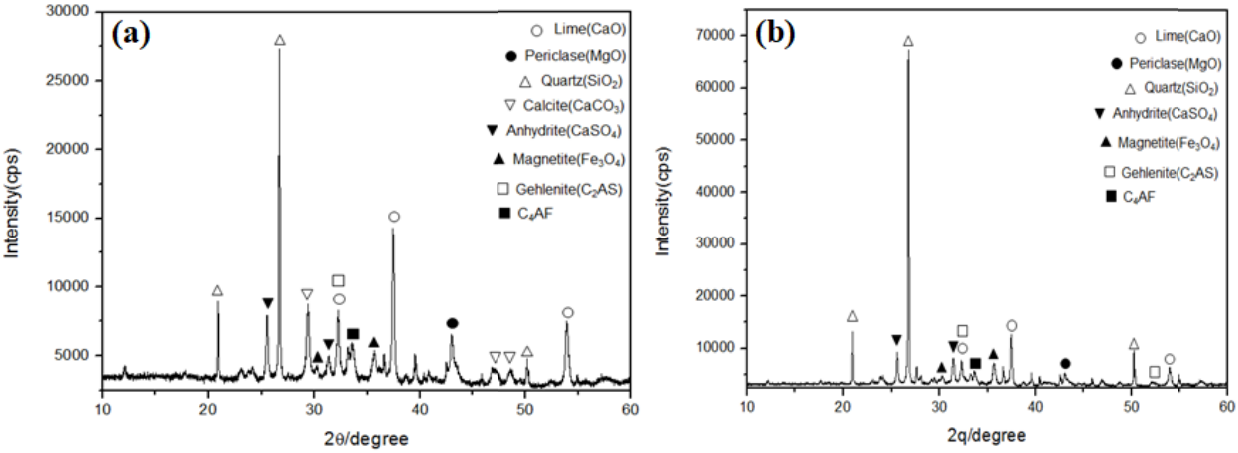

Fig. 5. (a) and (b) represented for CFBC fly ash and CFBC bottom ash.

Periclase 6.0%, Anhydrite 4.6%, Calcite 1.4%. Large amounts of CaO in the Samchuk green power circulating fluidized bed power plant.

3.3. Carbonation process of Heavy Metals and particulate matters

We studied the Samchuk green power circulating fluidized bed power plant coal ash samples by CO_2 immobilization and reported characteristics of those samples. The batch type reactor which we were used for this studies showed in the Figure 6. carbonation batch type reactor.

We optimize CO_2 immobilization technology for circulating fluidized bed coal ash and bottom ash samples, various conditions (water content, CO_2 concentration, temperature, pressure, etc.) to ensure optimum conditions and to control minerals or industrial byproducts. It is possible to stabilize harmful substances such as heavy metals by immobilizing CO_2.

$$CaO + H_2O \rightarrow Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \quad (6)$$

Figure. 6. Carbonation reactor-Batch type

In fig. 7 (a) and (b) shows CFBC coal ash before and after CO_2 immobilization, in these process, the hydrated lime (CaO) was decomposed and to form stabled calcite along with heavy metals was stabilized by carbonation process. The main advantages of CO_2 immobilization was, i) fixed harmful substances such as heavy metals by Capsulation effect (prevent secondary pollution) and ii) possible CO_2 immobilization in the power plant coal ash samples.

3.4. Effect of Solid/liquid ratio

We studied the effect of solid/liquid ratio of coal ash samples. In these process the fly ash was increased by about 3 times (S/L 0.1, S/L 0.2 and S/L 0.3) and measured the reaction time increased as shown in Fig. 8. In results show that the reaction is terminated quickly due to the low content of CaO components that can react with CO_2 in the fly ash and bottom ash samples.

The results indicated that the carbonate production efficiency was improved due to decrease of high liquid ratio (reduction of reaction time) and verified of relative occurrence of large amount of wastewater. The solid/liquid ratio was increased from 0.1 to 0.3, the reaction time will increase due to the high amount of CaO availability in both CFBC fly ash and bottom ash samples.

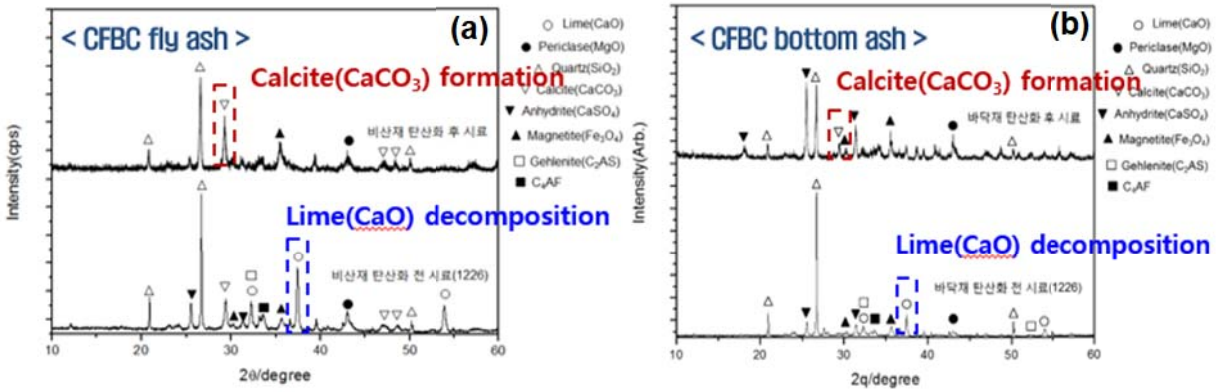

Figure. 7. (a) Carbonated CFBC fly ash, and (b) Carbonated CFBC bottom ash

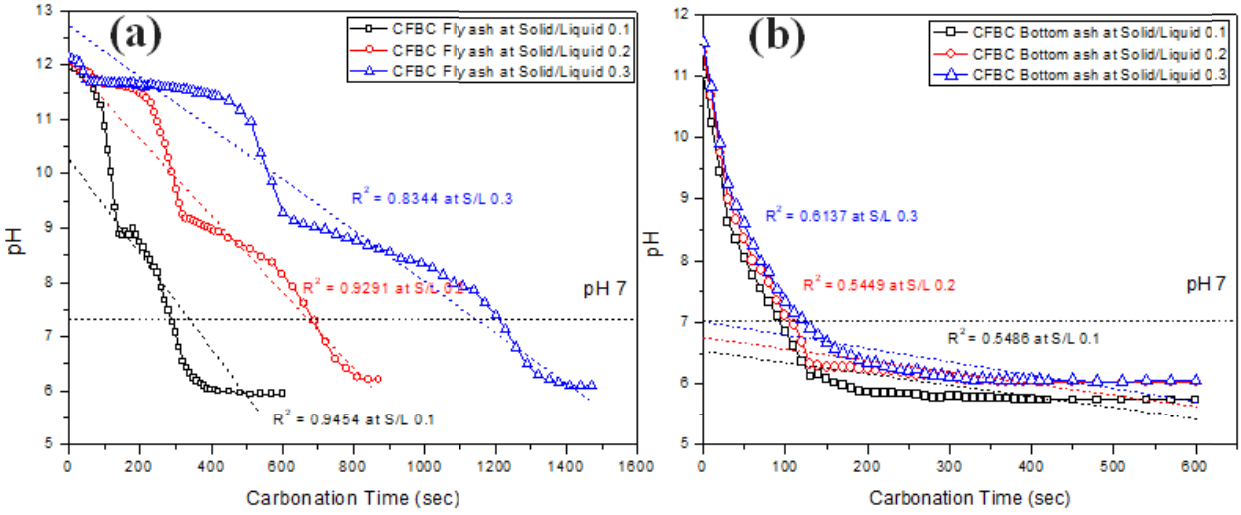

Figure. 8. (a) Carbonated CFBC fly ash (b) Carbonated CFBC bottom ash.

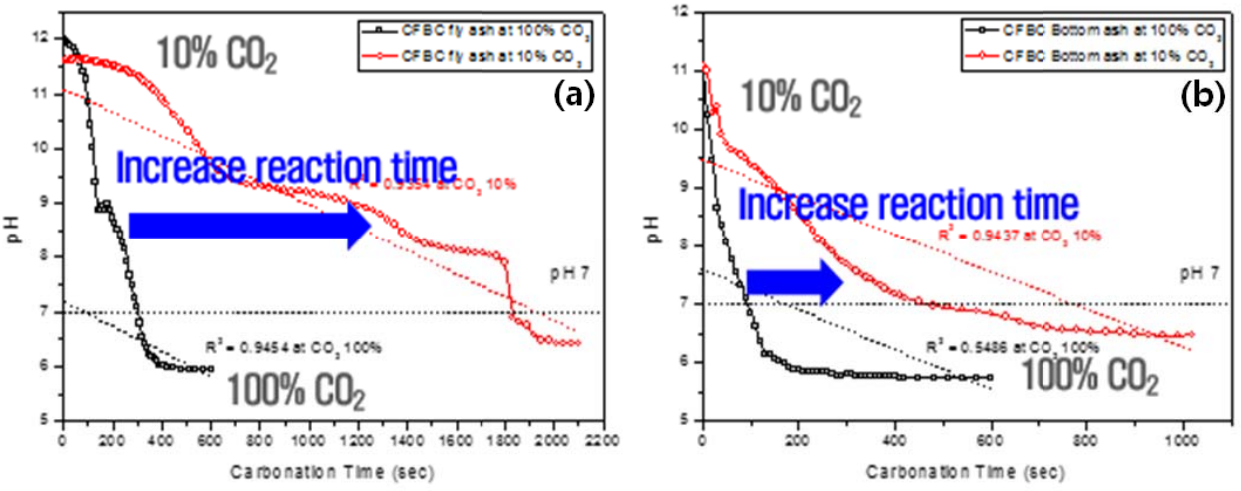

Figure. 9. (a) CFBC fly ash and (b) CFBC bottom ash samples.

3.5. Effect of CO_2 Concentration:

We studied two different CO_2 concentrations (10% CO_2 and 100% CO_2) for coal ash samples carbonation process. The results indicate the CO_2 immobilization reaction time increased for the production efficiency according to the concentration of 10% CO_2) gas supplied, but in the case of 100% CO_2 gas was supplied the reaction time is decreased for the CO_2 immobilization and increase the final stable solid production as shown in Fig.9 carbonation process with different CO_2 concentrations for CFBC fly ash and bottom ash samples. The CO_2 immobilization reaction time increase with actual exhaust gas CO_2 concentration (10%). Fly ash, 6 times increase reaction time and increase reaction time about 5 times as for bottom ash samples.

3.6. Heavy metals immobilization by carbonation process

Carbonation is highly effective immobilization of SOx, NOx and particulate matter etc.emissions from coal power plant. Assessment of heavy metal contents in CFBC (unit: mg/kg) as shown in Table. 6 heavy metals are presented in CFBC fly ash and bot-

Table 6. Heavy metal contents present in CFBC fly ash and bottom ash sample (unit: mg/kg)

Sample name	Pb	Cu	As	Cd	Cr	Zn	Hg
Fly Ash	9.0	32.3	7.5	5.6	31.1	92.8	0.589
Bottom Ash	2.1	7.9	25.0	4.8	30.7	80.5	0.007

Table 7: Leaching Characteristics of heavy metal contents in CFBC (unit: mg/kg)

Sample name	Pb	Cu	As	Cd	Cr	Hg
Fly Ash	0.00240	0.00082	0.00093	0.00020	0.01900	N.D
Bottom Ash	0.00200	0.00083	0.00100	0.00011	0.00130	N.D

Table 8: Immobilization of heavy metal by carbonation (unit: mg/kg)

Sample name	Pb	Cu	As	Cd	Cr	Hg
Fly Ash	0.00240	0.00082	0.00093	0.00020	0.01900	N.D
Fly Ash (After carbonation)	ND	ND	ND	ND	ND	N.D
Bottom Ash	0.00200	0.00083	0.00100	0.00011	0.00130	N.D
Bottom Ash (After carbonation)	ND	ND	ND	ND	ND	N.D

N.D. means non-detectable..

tom ash sample.

These dried CFBC fly ash and bottom ash was used for leaching experiment, after leaching process, Pb, Cu, As, Cd and Cr heavy metals was eluted in the leaching solution as shown in Table 7. leaching characteristics of heavy metal contents in CFBC (unit: mg/kg).

The CFBC fly ash and bottom ash samples was used to immobilization of heavy metals by carbonation process, these carbonated CFBC coal ash samples was used for leaching experiment, after leaching process, Pb, Cu, As, Cd, Cr and Hg heavy metals was not eluted in the leaching solution as shown in Table 8. After carbonation the heavy metal contents in CFBC (unit: mg/kg) samples, these results indicated that after carbonation process the heavy metals are stabilized and not eluted in the

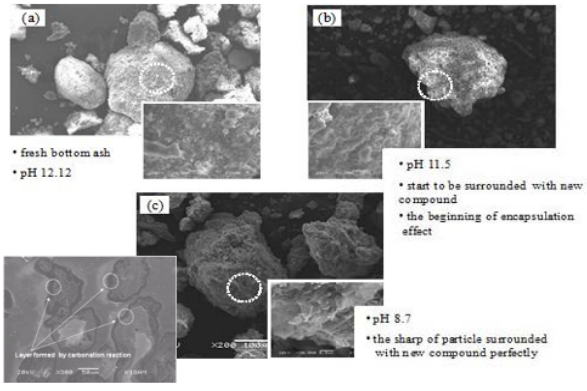

Figure. 10. Heavy Metals Encapsulation by Carbonation Process in CFBC coal ash sample.

leaching process and Fig.10 also indicated the SEM images for heavy metals encapsulation process by carbonation in CFBC coal ash sample.

Calcium carbonate ($CaCO_3$) is formed on the sur-

face of alkali waste particles by immobilization of CO_2. So that it can be stabilized so that heavy metals cannot be rereleased through encapsulation.

4. CONCLUSIONS:

There are several conventional methods for the removal of toxic heavy metals and particulate matters from coal power plant ash samples. Still, the complete extraction of the toxic species from waste and industrial effluents in order to reach acceptable levels represents a true challenge. We investigated the possibility of carbonation process for heavy metals removal and stabilization from coal ash samples and we confirmed carbonation is a good tool and definitely it's an emerging technology for toxic metals removal. CO_2 immobilization (accelerated carbonation) is a technology that can stabilize harmful substance such as heavy metals and particulate matters. Immobilization can stabilize heavy metals and particulate matter presented in coal ash samples through pH neutralization.

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