Monitoring of PM2.5 in coal ash samples & heavy metals stabilization by carbonation method

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ABSTRACT

Coal-fired 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. Their emissions threaten the health of people who live near these 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

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.

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].

Particulate matters widely dived into 3 categories 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.

Coal power plants are the major sources of pollutant emissions. Over 386,000 tons of 84 separate hazardous air pollutants released from over 400 cola power plants in 46 states of USA[3]. Currently the coal industries faces stringent emission regulations to limits the release of SO2, NOx, toxic volatile organic compounds, heavy metals, and particulate matter (PM). PM can contain any or all of the aforementioned chemical species or their compounds, plus water and biogenic organic species. PM2.5, or fine PM (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 where coal is the main energy source for power generation, such as China, India 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].

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

Country	**Time period**	**NOx,µg/m3**		**SOx,µg/m3**		**PM,µg/m3**	
		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

There are several reports published for the source of particulate matter emissions from industrial plants[5-10].

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. 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 respiratory illness[11-14]

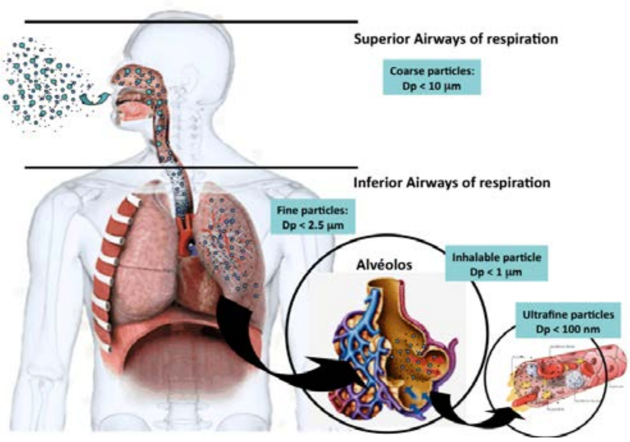


Figure.1. Health Impacts of Particulate Emissions [adopted from ref.15].

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 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].

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 μg/m3 (day) 500 μg/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	NO_2: 40 μg/m3 (year), NO_2: 200 μg/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	PM2.5 10 μg/m3 (year), PM10 20 μg/m3 (year) Directive 2001/80/EC: (monthly, total dust) 50 mg/m3 (old plants), 30 mg/m3 (new plants) Directive 2008/50/EC: 25 μg/m3 target PM2.5 (year), 50 μg/m3 (day) limit PM10, not to exceed on >35 days

	disease, stunted lung development (PM2.5), lung cancer; Cardiovascular: cardiac arrhythmias, acute myocardial infarction, congestive heart failure (PM2.5). Nervous system: ischemic stroke.	
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/m3 air; Directive 2004/107/EC: As 6ng/m3; Cd 5ng/m3; Ni 20ng/m3 (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 µg/m3 (air) Directive 2008/50/EC: 0,5 µg/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)

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].

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

3. Standard methods for sampling and measurement

The major methods are focusing on fine PM testing and measuring methods concentrate on determining 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.

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, SO_2, 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%

4. EXPERIMENTAL PROCEDURE

4.1 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 caol ash samples by using various mesh sizes of sieving machines. The flow sheet of procedure which we followed as per particle size distribution showed in Fig. 3

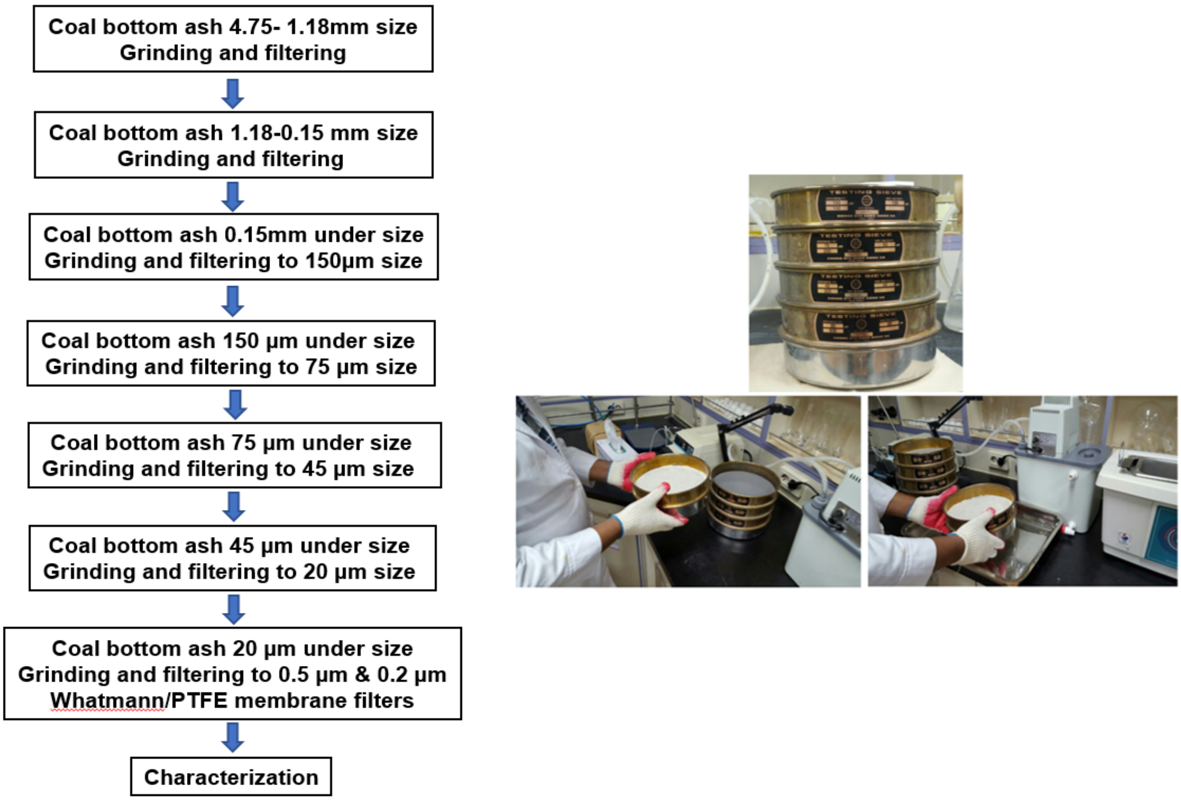


Figure 3. Experimental flow sheet for coal bottom ash sieving

The chemical compositions of coal ash sample is listed in Table 4.

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
	1652.7	99.99%	1713.5	99.99%	1641.9	99.99%	99.98
Total weight	1667.2	-14.5	1728.6	-15.1	1654.3		

Table 5. Composition of Coal ash samples

Collection period	Sample name	SiO2	Al2O3	Fe2O3	CaO	MgO	SO 3	Na2O	K2O	Etc	Ig-loss
16.9.21	F/A	33.50	16.70	26.80	13.90	5.76	-	0.04	1.20	2.10	2.99
	F/A	38.40	19.00	22.90	11.50	4.96	-	0.03	1.25	1.96	3.62
	B/A	68.60	9.30	8.68	6.68	2.92	-	0.09	2.96	0.77	0.28
16.11.8	B/A	40.00	20.90	11.30	18.20	6.49	-	0.03	1.51	1.65	2.47
	B/A	42.00	18.80	11.40	13.10	5.88	-	0.04	1.52	1.55	1.98
	F/A	35.60	22.70	19.70	21.20	7.13	-	0.02	1.12	1.88	1.99
16.12.3	F/A	36.90	16.30	15.80	27.60	7.94	-	0.03	0.99	1.65	3.15
16.12.6	F/A	26.40	13.40	17.20	33.30	10.00	-	0.02	0.87	6.57	5.06
16.12.9	F/A	22.40	13.60	11.90	32.40	9.16	-	0.02	0.89	7.89	6.40
16.12.18	F/A	21.20	11.40	17.60	23.10	7.54	-	0.02	0.81	7.41	6.15
16.12.21	F/A	27.70	14.70	20.50	25.00	7.54	-	0.01	0.89	5.56	4.00
17.1.16	F/A	31.40	16.80	11.70	26.70	9.89		0.02	0.98	6.56	4.60
17.1.16	F/A	29.86	16.61	10.37	27.07	9.89		0.02	1.00	5.09	3.98
17.2.2	B/A	23.99	16.61	10.85	29.01	8.11		0.02	0.99	5.11	4.12
17.2.2	B/A	39.46	9.46	6.20	35.31	11.25		0.06	1.25	4.80	4.00

Establishment of database through continuous monitoring of Samcheok circulating fluidized bed generation.

5 RESULTS AND DISCUSSION

We investigated the physical and chemical characteristics of Samchuk Green Power Circulating Fluidized Bed Power Plant coal ash samples. The CFBC bottom ash contains high amount of calcium because of the desulfurization process using limestone. The particle distribution is presented in Fig.4. The particles in between 0.15 ~ 1.18mm occupies about 80%.

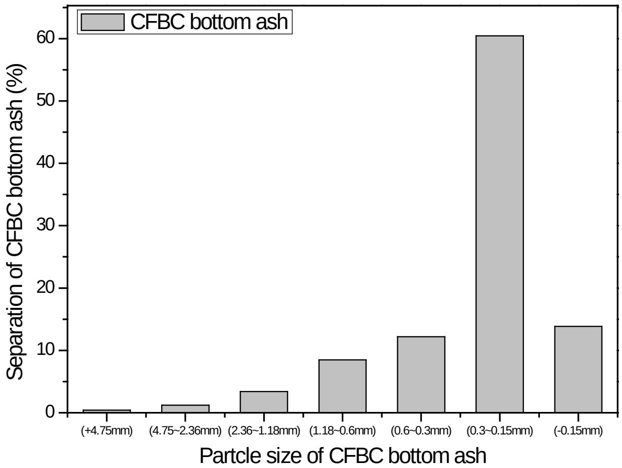
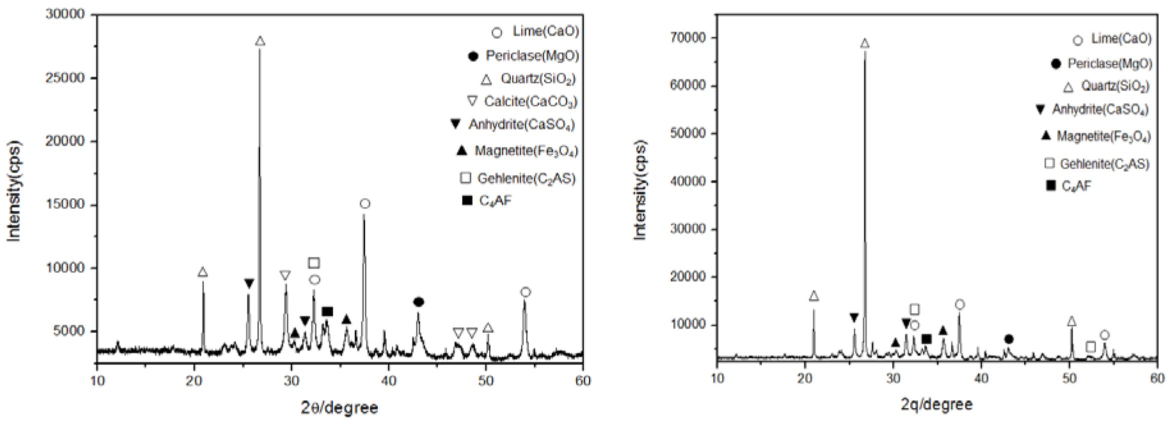


Figure.4 Particle size distribution of CFBC bottom ash



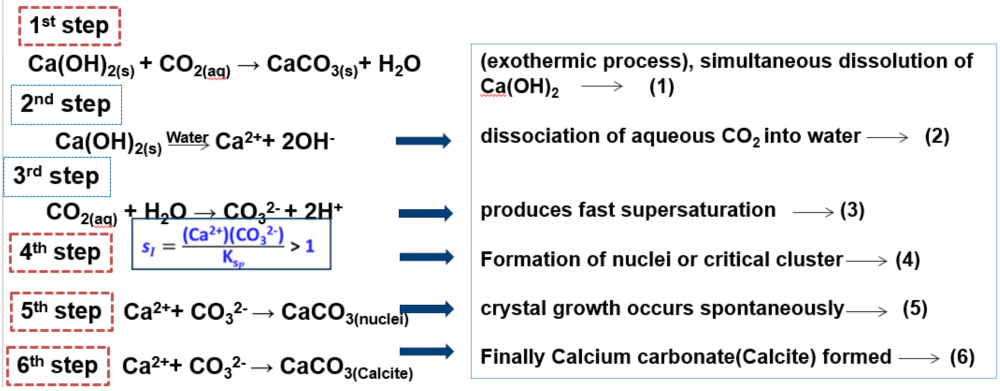
(a) (b)

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

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 37.0% and 30.6% of CaO is contained in fly ash and bottom ash, respectively.In fly ash, Lime 20.5%, Periclase 12.3%, Anhydrite 6.4%, Calcite 16.6% and in Flooring: Lime 9.9%, Periclase 6.0%, Anhydrite 4.6%, Calcite 1.4%. Large amounts of CaO in the Samchuk green power circulating fluidized bed power plant.

5.1Carbonation 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.



5.2 Carbonation process of Heavy Metals and particulate matters

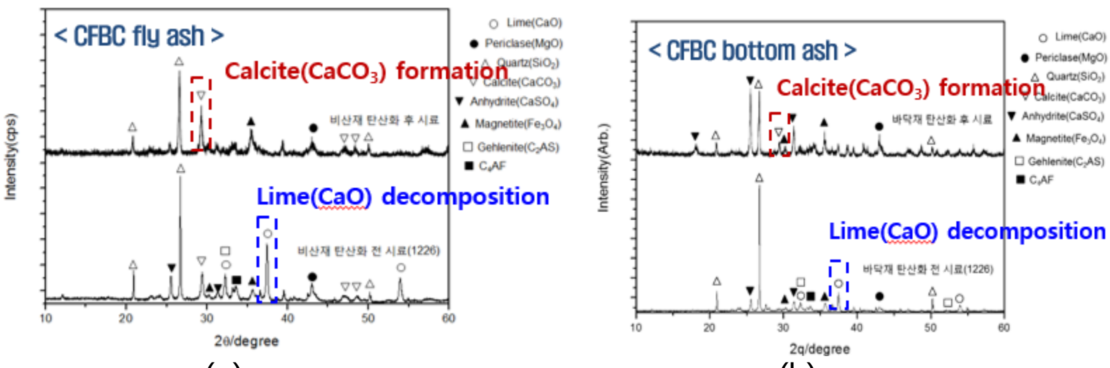
We studied the Samchuk green power circulating fluidized bed power plant coal ash samples by CO2 immobilization and reported characteristics of those samples. The batch type reactor which we were used for this studies showed in the Figure 6.

Figure. 6. Carbonation reactor-Batch type

We optimize CO2 immobilization technology for circulating fluidized bed coal ash samples, various conditions (water content, CO2 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 CO2.

$$\mathbf{CaO + H2O \rightarrow Ca\ (OH)\ 2 + CO2 \rightarrow CaCO3 + H2O}$$

Figure 7 (a) and (b) shows CFBC before and after CO2 immobilization Development of minerals. The advantages of CO2 immobilization is : 1.Fixed harmful substances such as heavy metals by Capsulation effect (prevent secondary pollution), 2. Possible CO2 immobilization in the power plant.



(a) (b)

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

5.3 Effect of Solid/liquid ratio

We studied the effect of solid/liquid ratio of coal ash samples. The correlation between carbonate production efficiency and wastewater generation according to power generation-H2O in the case of power plant CO2 immobilization (Fig.8).

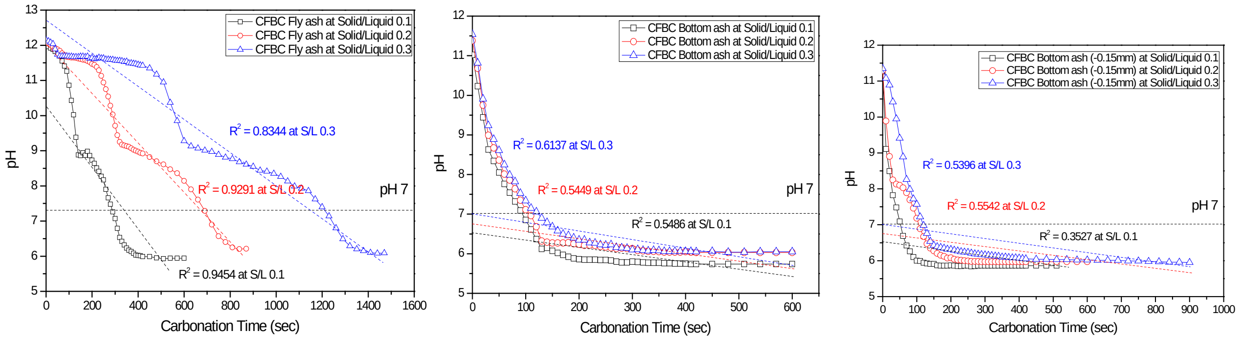


Figure. 7 (a) Carbonated CFBC fly ash (b) Carbonated CFBC bottom ash, (c) bottom ash with -0.15mm size.

The fly ash increased by about 4 times (S / L 0.1 → S / L 0.3) as reaction time increased although the reaction time is similar (small amount of CaO) (Fig.8).

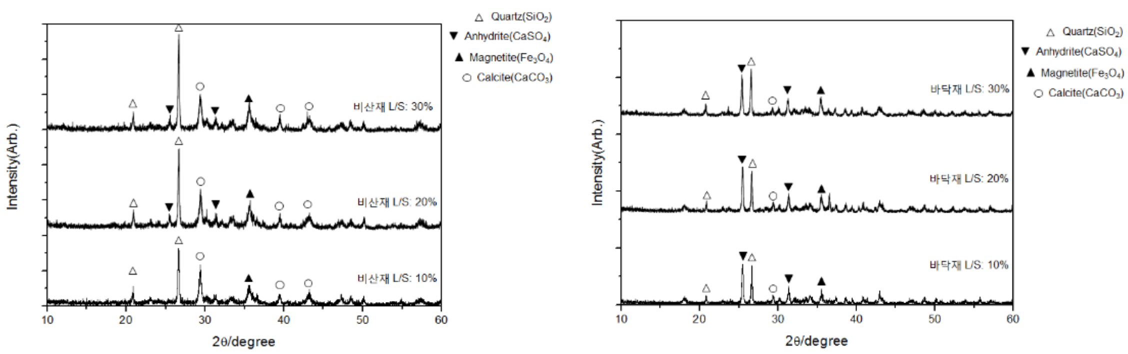


Figure. 8 (a) CFBC fly ash (b) CFBC bottom ash with different Liquid/solid ratios.

Improvement of production efficiency due to decrease of high liquid ratio (reduction of reaction time) - verified of relative occurrence of large amount of wastewater.

5.4 Effect of CO2 Conc.

We studied the effect of CO2 con for coal ash samples. We identify the production efficiency according to the concentration (about 10%) of the exhaust gas supplied when the power plant CO2 is fixed showed in Fig.9.

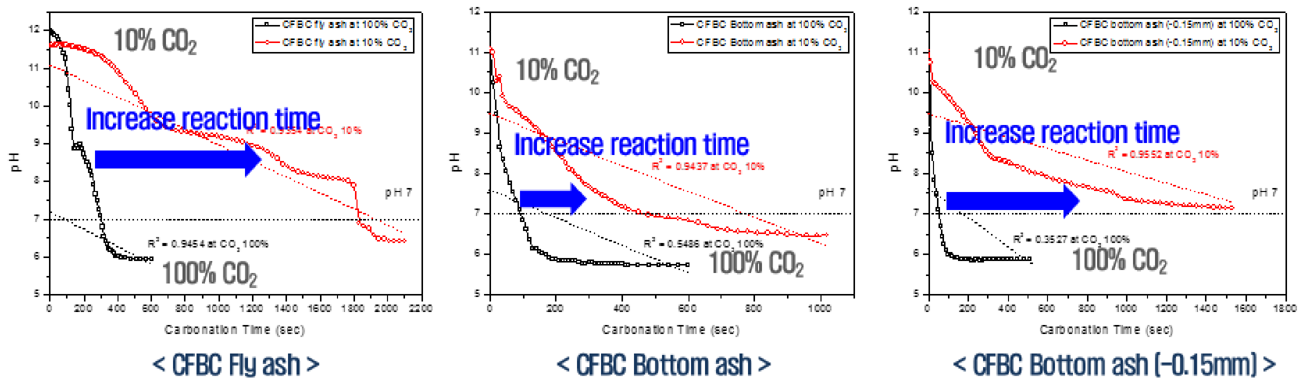


Figure. 9 (a) CFBC fly ash (b) CFBC bottom ash and (c) bottom ash with -0.15mm

CO2 immobilization reaction time increase with actual exhaust gas CO2 concentration (10%). Fly ash, 6 times increase reaction time, increase reaction time about 5 times as for bottom ash samples.

5.5 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) and leaching characterizes of heavy metals showed in Table 6 and Table 7. Table 6: Assessment of heavy metal contents in CFBC (unit: mg/kg). The immobilization of heavy metals by carbonation process are the below environmental standard values (Table 8). The heavy metals encapsulation by carbonation process showed in Fig.10.

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	N.D	N.D	N.D	N.D	N.D	N.D
Bottom Ash	N.D	N.D	N.D	N.D	N.D	N.D
Fly Ash	0.00240	0.00081	N.D	N.D	N.D	N.D
Bottom Ash	0.00038	0.00083	0.00018	0.00011	0.00130	N.D
Standard	3.0	3.0	1.5	0.3	1.5	0.005

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

Sample name	Pb	Cu	As	Cd	Cr	Hg
Fly Ash(After carbonation)	0.00029	0.00082	0.00093	0.00020	0.01900	N.D
Bottom Ash(After carbonation)	0.00200	0.00060	0.00100	ND	0.00026	N.D
Environmental Standard Value	3.0	3.0	1.5	0.3	1.5	0.005

N.D. means non detectable, and the detection limit of sample is 0.0001 mg/L.

We achieved the heavy metals concentrations by immobilization of carbonation. Those values are under environmental standard values by domestic waste dissolution method (KCLP).

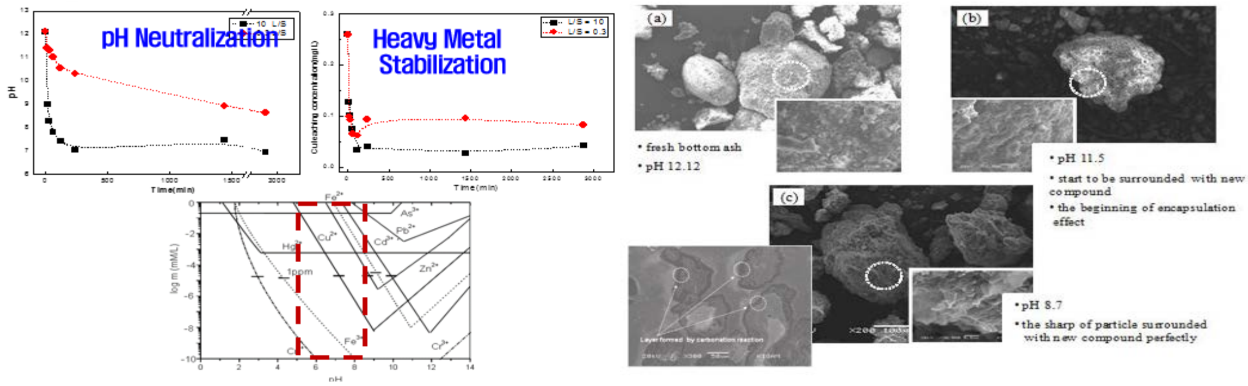


Figure. 10 Heavy Metals Encapsulation by Carbonation Process.

Calcium carbonate ($CaCO_3$) is formed on the surface of alkali waste particles by immobilization of CO2. So that it can be stabilized so that heavy metals cannot be rereleased through encapsulation.

6 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 its an emerging technology for toxic metals removal. CO2 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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