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Systematic investigation of heavy metals from MSWI fly ash and bottom ash samples

Ramakrishna. CH[1], Thriveni. T[2] and Ahn Ji Whan[1*]
[1,2]Hanil Cement, 302 Maepo-gil, Maepo-eup, Danyang-gun, Chungcheongbuk-do, Korea
[1*]Mineral Processing Division, Korea Institute of Geoscience and Mineral Resources (KIGAM), Gwahagno-124, Yuseongu-gu, Daejeon, Korea

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Abstract

Disposal of municipal solid waste has become a major problem in many countries around the world. As landfill space for the disposal of ash from Municipal Solid Waste Incineration (MSWI) becomes scarce, numerous reports and researches address the various environmental issues about the municipal solid waste incineration waste management and other particulate matters with the range of 10 ~ 2.5. Although in many developing and industrialization countries landfill with the disposal of municipal solid waste, open incineration has become a common practice. Large municipal waste incinerators are major industrial facilities and have the potential to be significant sources of environmental pollution. Despite the significant volume reduction from incineration, waste recycling is important to ensuring the future welfare of mankind. The main goal of the present work is the physical and chemical characterization of the local incineration bottom ash towards its eventual re-utilization. In this paper, we reported the studies on physical and chemical characteristics of municipal solid waste incineration (MSWI) fly ash and bottom ash containing particulate matter whose particulate sizes are lower than PM_{10}, $PM_{2.5}$ and heavy metal were investigated.

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

1. INTRODUCTION

Municipal solid waste (MSW) is an inevitable by-product of civilization and a prime target for clean technology innovation. It is threat to public health and environmental security and a strategic renewable resource. This solid waste generated globally is expected to grow from 1.5 billion tons in 2014 to 2.2 billion tons by 2023. 97% of waste volume growth is expected to come from Asia Pacific and Africa, primarily in developing economies across these regions. The incineration of municipal solid waste is an alternative method for sustainable environment [1].

On a global scale, calculating the amount of waste generated is challenging because many countries do not track waste generation or disposal statistics. World Bank estimates that roughly 3 billion urban residents generated an average 1.2 billion kg per capita per day (1.3 billion tonnes per year or 1.43 billion tonnes per year) in 2012. By 2025, this is expected to increase to 4.3 billion urban residents generating about 1.42 kg per capita per day of MSW (2.2 billion tonnes per year or 2.4 billion tonnes per year). This represents a 900 million tonnes (992 million tonnes) increase in a little over a decade, a near doubling of the total volume of MSW generated globally today [2].

Municipal Solid Waste (MSW) Management is becoming an integral part of the urban environment and planning of the modern infrastructure to ensure

†To whom corresponding should be addressed.
Tel : +82-00-00 E-mail :

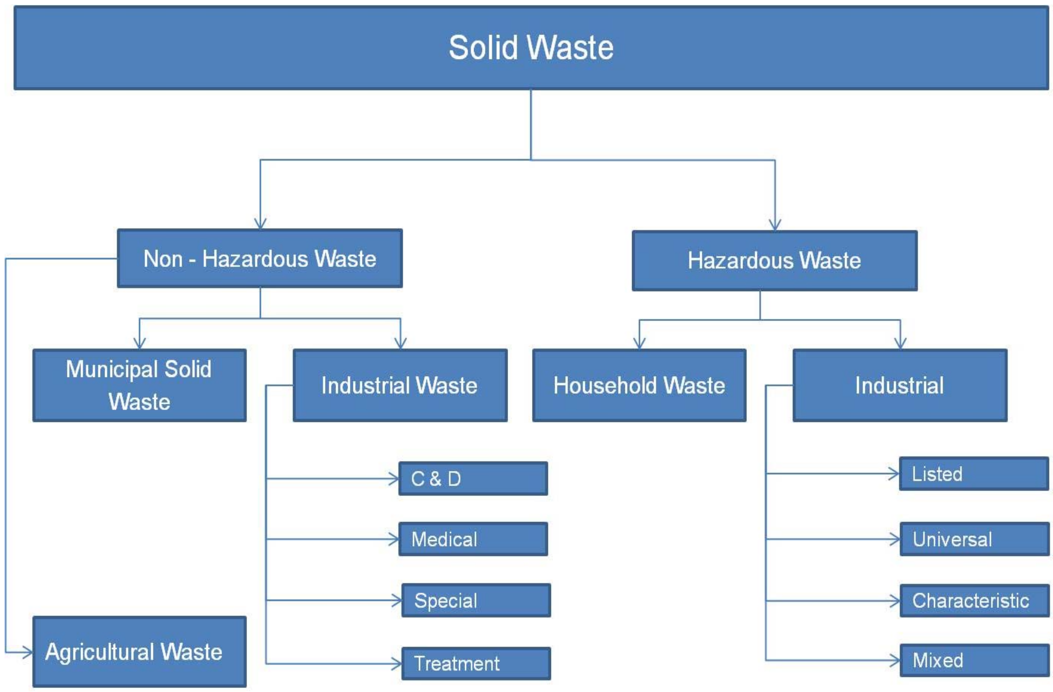

Fig. 1. Solid waste category tree

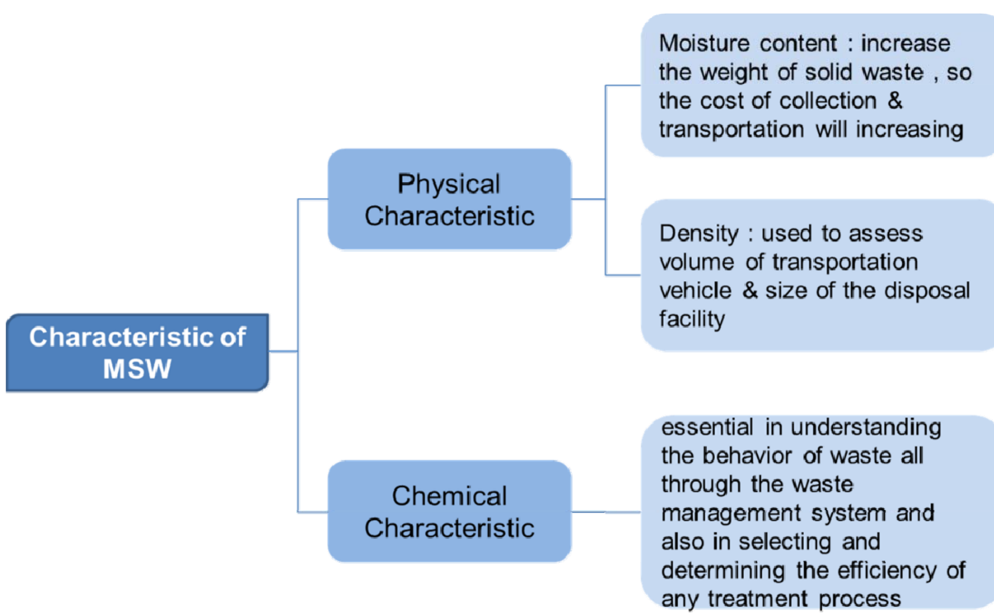

Fig. 2. Basic characteristics of municipal solid waste

a safe and healthy human environment while considering the promotion of sustainable economic development. In developing countries such as China, India, Malaysia, Thailand and Bangladesh severe municipal solid waste management problems are reported [3-8].

Recently, there are several comprehensive reports and publications are available are about global municipal solid waste generation [9-12]. MSWI ash can

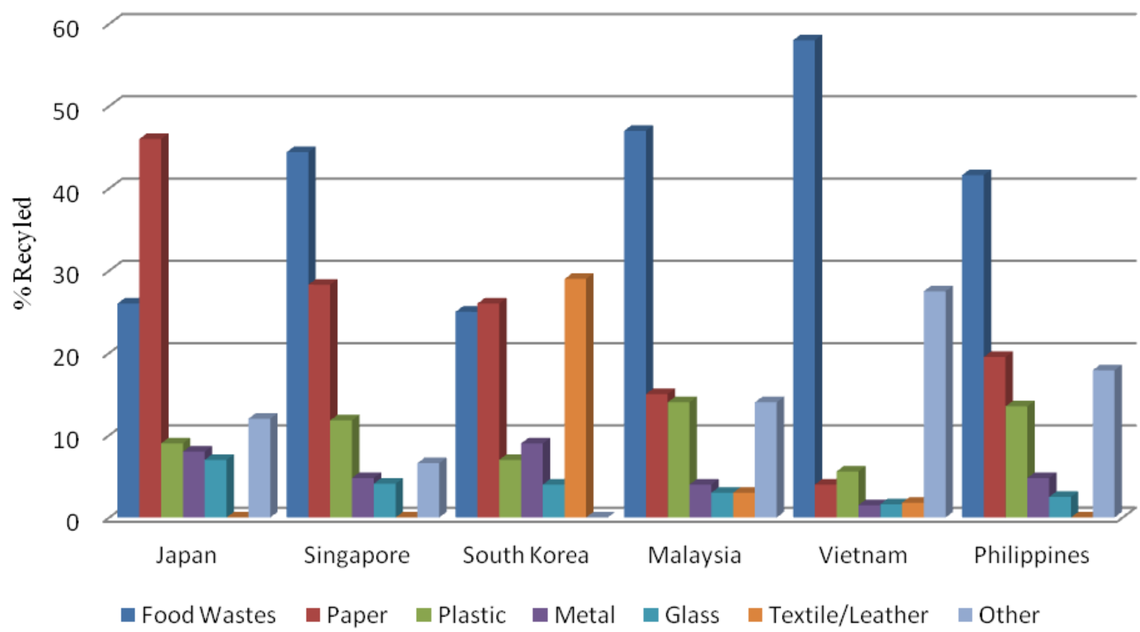

Fig. 3. MSW composition from Asia

be classified into two classes: one is bottom ash that occupies 90 % of the total MSWI ash and contains glassy materials, ceramics, mainly ferrous materials; and the other is fly ash that has been already treated by landfill. Therefore, the development of a feasible method that enables MSWI bottom ash to be used as alternative resources is urgent. MSWI bottom ash can be utilized as aggregate substitutes in the construction field. However, toxic heavy metal ions contained in bottom ash can be easily dissolved in aqueous solutions, and that presents severe environmental problems, and prevents bottom ash being reused.

Generally, the term solid waste refers to non-hazardous waste. But according to the Resource Conservation and Recovery Act (RCRA) and other state regulations, hazardouswaste is also a part of solid waste. Solid waste tree showed in Fig.1, based on resource conservation and recovery act, US EPA and also the characteristics of a municipal solid waste is presented in Fig.2.

The disposal of inorganic waste presents logistical, financial and environmental issues. Technologies that enable the wastes to be utilized as alternative resources for the synthesis of useful materials and can reduce the hazardous properties of wastes are urgently required. The increasing CO_2 content and research on the capture and long term effect on the climate has increased global interest and research on the capture and longtime storage of CO_2.

The municipal generation by country (high income and lower middle income including) and their current data and projections for 2025 [2], showed in Fig.3. The proportion of recyclables (paper, plastics) is high in the developed countries. The low proportion of recyclables in developing economies can be attributed to the market value of recyclables.

In this paper, we reported the characteristics of municipal solid waste (MSWI) bottom ash produced in South Korea were investigated and studied the feasible treatments to the MSWI samples to stabilize heavy metal ions before recycling. The hazardous materials can be stabilized by accelerated carbonation technology. When carbonates are quickly synthesized by accelerated carbonation, especially on the surface of bottom ash, heavy metal ions are solidified. We have applied accelerated carbonation technology to the immobilize heavy metals and bottom ash.

2. EXPERIMENTAL PROCEDURE

2.1. MATERIALS AND SAMPLE PREPARAITON

In this study, we collected Seoul municipal solid waste incineration bottom ash samples from four different areas. After receiving the samples we dried the samples at room temperature for the removal of moisture from that samples. After drying, we sieved the bottom ash samples by using various mesh sizes of sieving machines, Table 1, shows MSWI bottom

Table 1. Seoul municipal solid waste incineration bottom ash distribution rate by particle-size

Sieving machine size (mm)	Gang-nam resource recovery		No-Won resource recovery		Ma-pho resource recovery		Yang-Cheon resource recovery	
	Sample weight	Distribution rate	Sample weight	Distribution rate	Sample weight	Distribution rate	Sample weight	Distribution rate
1st iron separation	3.30	4.02	4.40	2.99	12.50	6.02	4.25	2.86
2nd iron separation	4.00	4.88	10.15	6.91	31.45	15.16	6.05	4.06
4.75mm over	24.10	29.39	20.90	14.23	57.20	27.57	18.25	12.26
4.75~2.36	11.60	14.15	24.55	16.71	36.65	17.66	17.45	11.72
2.36~1.18	11.95	14.57	23.35	15.90	31.70	15.28	32.50	21.83
1.18~0.6	9.45	11.52	15.55	10.59	16.65	8.02	29.60	19.89
0.6~0.3	5.85	7.13	9.55	6.50	7.40	3.57	15.60	10.48
0.3~0.15	7.05	8.60	20.2	13.75	7.20	3.45	14.00	9.41
0.15 under	4.70	5.73	18.25	12.42	6.75	3.25	11.15	7.49
Total	82.0 (kg)	99.99 (%)	146.90 (kg)	100.0 (%)	207.50 (kg)	99.98 (%)	148.85 (kg)	100.0 (%)

Table 2.1. Chemical composition of Yang-Cheon bottom ash samples.

Yang-Cheon bottom ash sample										
Particle size (mm)	SiO_2	Al_2O_3	CaO	MgO	P_2O_5	Na_2O	Fe_2O_3	K_2O	TiO_2	Cl	Ig loss
10.0mm/4.75 (4mesh)	45.4	7.44	21.9	1.6	6.74	6.25	1.59	1.42	0.36	0.44	4.06
4.75/2.36 (4-8mesh)	26.7	9.18	30.8	1.43	11.0	3.33	2.14	1.92	0.63	1.06	8.12
2.36/1.18 (8-16mesh)	21.4	10.6	33.3	1.21	4.47	3.09	2.71	2.02	0.93	1.60	15.5
1.18/0.6 (16-30mesh)	15	9.75	38.2	1.15	4.20	2.91	2.36	1.64	1.05	1.92	19.2
0.6/0.3 (30-50 mesh)	11.4	9.91	40.0	1.20	4.88	2.54	2.31	1.39	1.05	2.10	20.5
0.3/0.15 (50-100mesh)	8.47	10.1	40.3	1.25	4.82	2.49	2.21	1.09	1.16	2.16	23.3
0.15/0.075 (100-200mesh)	7.06	9.94	39.6	1.28	3.0	2.40	1.95	0.96	1.13	2.08	28.1

Table 2.2. Chemical composition of Gang-nam bottom ash samples.

Gang-nam bottom ash sample											
Particle size (mm)	SiO_2	Al_2O_3	CaO	MgO	P_2O_5	Na_2O	Fe_2O_3	K_2O	TiO_2	Cl	Ig loss
10.0mm/4.75 (4mesh)	47.0	10.6	19.0	1.54	4.74	6.05	2.07	1.62	0.60	0.39	3.69
4.75/2.36 (4-8mesh)	28.4	11.4	29.7	1.35	8.61	3.15	2.96	1.87	0.88	0.73	8.49
2.36/1.18 (8-16mesh)	28.5	11.6	29.0	1.30	6.57	2.55	3.20	2.08	1.0	0.78	10.4
1.18/0.6 (16-30mesh)	17.4	11.5	36.7	1.24	6.52	2.29	3.44	1.64	1.12	1.18	14.5
0.6/0.3 (30-50 mesh)	13.2	11.6	38.3	1.21	6.17	2.04	3.33	1.25	1.14	1.37	18.1
0.3/0.15 (50-100mesh)	8.63	12.0	40.4	1.11	4.18	1.79	2.75	0.85	1.07	1.56	23.5
0.15/0.075 (100-200mesh)	6.98	12.2	41.1	1.02	2.63	1.49	2.08	0.85	0.90	1.62	27.2

Table 2.3. Chemical composition of No-Won bottom ash samples.

No-Won bottom ash sample											
Particle size (mm)	SiO_2	Al_2O_3	CaO	MgO	P_2O_5	Na_2O	Fe_2O_3	K_2O	TiO_2	Cl	Ig loss
10.0mm/4.75 (4mesh)	40.8	7.15	23.9	1.46	9.51	6.96	0.27	1.45	0.48	0.33	3.53
4.75/2.36 (4-8mesh)	29.2	7.98	30.5	1.43	11.0	4.22	2.92	1.78	0.85	0.66	7.45
2.36/1.18 (8-16mesh)	24.5	10.4	31.7	1.44	7.03	3.93	3.12	2.15	1.15	0.99	11.5
1.18/0.6 (16-30mesh)	15.0	11.2	36.4	1.4	6.04	3.32	3.04	1.69	1.40	1.31	16.9
0.6/0.3 (30-50 mesh)	11.1	12.2	37.8	1.29	5.23	2.88	2.76	1.27	1.46	1.54	20.5
0.3/0.15 (50-100mesh)	6.35	12.5	39.3	1.06	3.20	2.37	1.94	0.87	1.13	1.83	27.5
0.15/0.075 (100-200mesh)	4.94	12.2	38.5	0.80	1.59	2.03	1.20	0.71	0.73	1.91	33.8

ash distribution rate by particle-size was listed.

After sieving the different sizes bottom ash samples, the carbonation process was conducted to stabilize the high content of hazardous heavy metals in MSWI bottom ash samples. The carbonation 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.

3. RESULTS AND DISCUSSION

3.1. Characteristics of MSWI bottom ash samples:

X-ray fluorescence (XRF) analysis of the chemical compositions for different MSWI bottom ash samples were presented. Yang-Cheon bottom ash samples shows in the Table 2.1, Gang-Nam bottom ash samples shows in Table 2.2, No-Won bottom ash samples shows in Table 2.3 and Ma-pho bottom ash samples shows in table 2.4.

Table 2.1 shows the chemical composition of Yang-Cheon- MSWI bottom ash major components are 20 to 40% of CaO, 10 to 40% of SiO_2 and 7-10% Al_2O_3 are present in different particle size of bottom ash samples and less amount of 1-1.5% MgO, 3-6% P_2O_5 and other elements are present in different MSWI bottom ash samples.

Table 2.2 shows the chemical composition of Gang-nam MSWI bottom ash major components are 19 to 41% of CaO, 10 to 47% of SiO_2 and 10-12% Al_2O_3 are present in different particle size of bottom ash samples and less amount of 1-1.5% MgO, 3-8% P_2O_5 and other elements are present in different MSWI bottom ash samples.

Table 2.3 shows the chemical composition of No-Won, MSWI bottom ash major components are 23 to 39% of CaO, 10 to 40% of SiO_2 and 7-12% Al_2O_3 are present in different particle size of bottom ash samples and less amount of 1-1.4% MgO, 1-10% P_2O_5 and other elements are present in different MSWI bottom ash samples.

Table 2.4 shows the chemical composition of Ma-pho MSWI bottom ash major components are 20 to 43% of CaO, 10 to 45% of SiO_2 and 10-12% Al_2O_3 are present in different particle size of bottom ash samples and less amount of 1-1.6% MgO, 2-7% P_2O_5, 2-3% Fe_2O_3 and other elements are present in different MSWI bottom ash samples.

In tables 2.1-2.4, are shows the chemical composition of MSWI Bottom ash was mainly composed by SiO_2, CaO, and Al_2O_3. But, the MSWI bottom ash also contain the plenty of heavy metals and chlorine. The heavy metals and chlorine included in bottom ash limited for its use, when recycle the bottom ash as an aggregate. Because there are serious problems of secondary environment pollution due to toxic component. Therefore, the stabilization of heavy metals and the removal of chlorine in the bottom ash

Table 2.4. Chemical composition of Ma-pho bottom ash samples.

Ma-pho bottom ash sample											
Particle size (mm)	SiO_2	Al_2O_3	CaO	MgO	P_2O_5	Na_2O	Fe_2O_3	K_2O	TiO_2	Cl	Ig loss
10.0mm/4.75 (4mesh)	45.8	10.4	20.3	1.64	4.65	6.17	3.43	1.34	0.84	0.26	3.5
4.75/2.36 (4-8mesh)	31.5	11.4	27.9	1.36	7.63	3.39	3.46	1.71	1.05	0.50	7.6
2.36/1.18 (8-16mesh)	26.4	12.0	30.4	1.39	6.15	2.69	3.72	1.71	1.25	0.64	11.1
1.18/0.6 (16-30mesh)	17.4	11.4	37.1	1.30	5.76	1.95	3.79	1.40	1.41	0.82	14.9
0.6/0.3 (30-50 mesh)	10.6	10.5	41.4	1.29	5.26	1.49	3.31	0.09	1.38	0.95	20.1
0.3/0.15 (50-100mesh)	8.61	10.1	42.2	1.29	4.63	1.29	3.18	0.73	1.44	0.96	22.8
0.15/0.075 (100-200mesh)	5.55	9.44	43.8	1.15	2.35	0.95	2.22	0.50	1.11	0.99	29.2

Table 3. Heavy Metal contents in MSWI bottom ash samples

Element (mg/kg)	EU	Japan	U.S.A	Korea
As	0.12-190	6.5	17.9-189	46.3-151.2
Cd	0.3-71	13.0	2.62-70.6	0.0-41.2
Cu	190-8,200	2,700	1,140-6,980	972.4-4,235
Cr	23-3,200	<0.7	86-2,005	179.9-1,496
Hg	0.02-7.8	0.19	0.0-2.25	0.0-3.194
Pb	98-14,000	1,100	1,200-6,600	229.4-3,145
Cl	800-4,200	14,000	1,105-4,190	26,000-30,000

are very important process to recycle bottom ash as alternative aggregate.

3.2. Heavy metals in different MSWI bottom ash samples and leaching concentrations:

The MSWI bottom ash samples having different heavy metals such as As, Cd, Cu, Cr, Hg, Pb and Cl in different countries reported in the Table 3. In order to evaluate the environmental stability of MSWI bottom ash, heavy metal content and leaching test were conducted.

We investigated the leaching concentrations of heavy metals in different municipal solid waste ash samples. The heavy metals content is reported in mg/kg in various bottom ash samples with various particles sizes and the leaching characteristics of heavy metals in bottom ash samples also reported in the Table 4.1 for Gang-nam bottom ash samples and Table 4.2 for Ma-pho bottom ash samples.

In Table 4.1, shows the heavy metal concentrations present in Gang-nam MSWI bottom ash sample in various particle sizes, these results indicated that the heavy metals such as Cu and Pb have high concentrations (>2000ppm), and these metals concentrations is high in <0.15mm size of particle containing bottom ash sample. It suggested that heavy metals concentrations is more in fine particles (<0.15mm) of MSWI bottom ash samples. After leaching process less amount such as 0.33mg/L for Cr, 6mg/L for Cu, 0.07mg/L for As and 0.1mg/L for Pb metals are eluted, reaming high concentrated metals are not eluted from MSWI bottom ash samples by these leaching process.

In Table 4.2, shows the heavy metal concentrations present in Ma-pho MSWI bottom ash sample in various particle sizes, in these results also indicated that the heavy metals such as Cu and Pb have high concentrations than Cr and As. These metals concentrations is high in <0.15mm size of particle containing bottom ash sample. It suggested that heavy metals concentrations is more in fine particles (<0.15mm) of MSWI bottom ash samples. In leaching process the metals, 0.29 mg/L for Cr, 5.86 mg/L for Cu, 0.08 mg/L As and 0.09 mg/L Pb was eluted. These eluted concentration of metals from MSWI bottom ash samples were stabilized by carbonation process.

3.3 Leaching Concentrations of Heavy metals with carbonation.

We are using MSWI Gang-nam bottom ash (<0.5mm size) samples for carbonation process 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^oC temperature and the carbonation process was termi-

Table 4.1. Heavy metals content and the leaching characteristics of heavy metals in Gang-nam bottom ash samples

Heavy metals present in Gang-nam bottom ash sample								
Particle size (mm)	Cr		Cu		As		Pb	
	Content (mg/Kg)	Leaching conc. (mg/L)	Content (mg/Kg)	Leaching conc. (mg/L)	Content (mg/Kg)	Leaching conc. (mg/L)	Content (mg/Kg)	Leaching conc. (mg/L)
10.0mm/4.75 (4mesh)	150.3	ND	1200.7	1.55	<0.5	ND	205.4	ND
4.75/2.36 (4-8mesh)	210.7	ND	1300.8	2.75	3.5	ND	1457.5	0.01
2.36/1.18 (8-16mesh)	286.0	0.13	2500.0	3.26	17.5	0.05	1751.2	0.08
1.18/0.6 (16-30mesh)	380.7	0.21	3324.5	4.26	18.5	0.06	1822.7	0.08
0.6/0.3 (30-50 mesh)	356.4	0.29	2600.7	5.43	20.2	0.06	2150.0	0.09
0.3/0.15 (50-100mesh)	313.7	0.32	2757.7	5.93	21.5	0.07	2102.5	0.10
0.15/0.075 (100-200mesh)	331.5	0.33	2956.9	6.00	239.8	0.07	2200.2	0.10

Table 4.2. Heavy metals content and the leaching characteristics of heavy metals in Ma-pho bottom ash samples

Heavy metals present in Ma-pho bottom ash sample								
Particle size (mm)	Cr		Cu		As		Pb	
	Content (mg/Kg)	Leaching conc. (mg/L)	Content (mg/Kg)	Leaching conc. (mg/L)	Content (mg/Kg)	Leaching conc. (mg/L)	Content (mg/Kg)	Leaching conc. (mg/L)
10.0mm/4.75 (4mesh)	337.5	ND	1179.4	2.3	<0.5	ND	927.5	ND
4.75/2.36 (4-8mesh)	347.2	0.05	2700.9	3.10	90.0	ND	1211.8	ND
2.36/1.18 (8-16mesh)	448.3	0.15	4521.9	4.41	108.8	0.05	1436.7	0.05
1.18/0.6 (16-30mesh)	414.6	0.20	3381.8	4.96	117.8	0.08	1487.3	0.07
0.6/0.3 (30-50 mesh)	481.8	0.27	3197.4	5.86	109.9	0.05	1282.3	0.06
0.3/0.15 (50-100mesh)	582.0	0.29	2607.8	5.43	137.1	0.06	1242.8	0.09
0.15/0.075 (100-200mesh)	548.5	0.21	2885.2	4.26	237.2	0.06	1464.2	0.08

nated when the measured pH was below 7 and sample was filtered and dried at 80^{o}C for 12h duration time. These dried carbonated MSWI bottom ash was used for leaching experiment, after these leaching

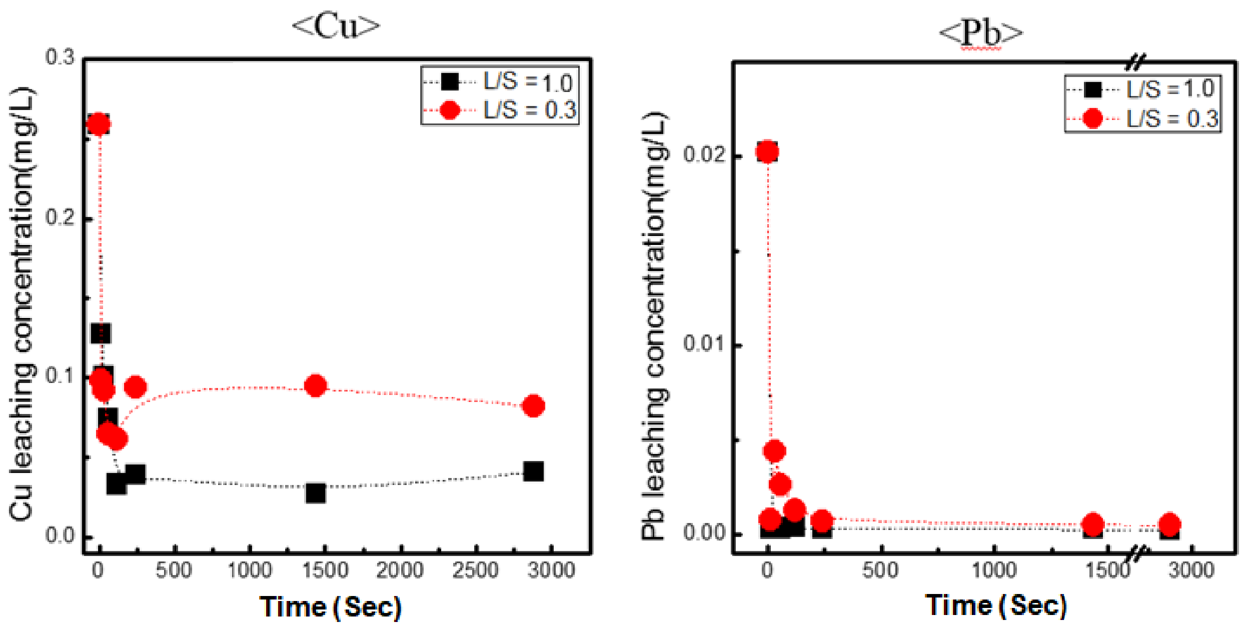

Figure 4. Effect of carbonation on leaching concentrations of heavy metals

process, Cu and Pb heavy metals is not eluted in the leaching solution results are shown in Fig. 4. These results indicated that after carbonation process by using the liquid and solid ratio is 0.3 and 1.0. Both copper and lead are effectively stabilize with the carbonation process.

4.CONCLUSIONS

The accelerated carbonation of solid wastes containing alkaline minerals such as Ca and Mg before their landfill treatment is effective for decreasing the mobility of heavy metals by adjusting pH to below 7 at which their solubility is lower. Worldwide trend towards encouraging carbonation treatment for waste alkaline solids emitted from various sources such as MSWI ash, coal ash and mining residues etc. Our research results shows the carbonation studies using MSWI bottom ash and CO_2 in order to increase the recycling percentage of the ash and reduce the concentration of atmospheric CO_2 in Korea. Carbonation is an ecofriendly process for the removal of heavy metals municipal solid waste bottom ash samples.

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