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COMBUSTION CHARACTERISTICS AND EMISSION OF HAZARDOUS AIR POLLUTANTS IN COMMERCIAL FLUIDIZED BED COMBUSTORS FOR SEWAGE SLUDGE

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ABSTRACT

Since the disposal of sewage sludge in ocean has been prohibited recently according to London Dumping Convention, technological need for treating sewage sludge safely and efficiently are getting increased in Korea. FBC (Fluidized Bed Combustor) technology has been selected and utilized as one of the alternatives because of combustible content in sludge, on-going process development to maintain the best combustion efficiency, and good heat recovery for energy utilization. In this paper, the process and combustion characteristics of commercially operating FBC incineration plants with the capacity ranging from 50 – 150 tons of sludge per day were investigated by comparing emission data from 4 different plants. Concentrations of hazardous gaseous pollutants (HAPs) such as fine particulate matter, heavy metals, and dioxin from sludge combustion before and after air pollution control devices were measured and analyzed at commercial operating conditions of one typical incinerator. Most of emission data at stack showed under the environmental regulatory limits. Mercury and some heavy metals emission have been reduced significantly as co-beneficial effect since the air pollution control configuration was well arranged and installed to control the regulatory gases such as NOx, SO_2, particulates, and dioxin. Sludge, especially generated from industrial plants, contained measurable amounts of acidic materials and heavy metals including Hg. Therefore, such metal emission should receive an attention by monitoring them and further mass balance study for better understanding their fates in the process must proceed in future.

Keywords : Sewage Sludge, Fludized Bed Combustor, Hazardous Air Pollutants, Particulate Matter, Heavy Metals, Mercury, Air Pollution Control Device

INTRODUCTION

FBC technology has been widely used for sewage sludge combustion. In the USA, application of fluidized beds for the incineration of municipal sewage sludge began in the early 1960s. In Europe, the first fluidized bed sludge incinerator was installed in 1964 in Germany for the combustion of refinery sludge. And one year after another incinerator was installed in Switzerland for the combustion of municipal sewage sludge (Loll et al., 1996). Since then, the use of FBC for sludge combustion has been expanded very fast. The success of FBC technology for sewage sludge

combustion has been attributed to following advantages (Muellen et al, 1992). First, the intimate mixing in the bed with its resulting high turbulence and the large surface area of the inert bed material available for heat transfer provide virtually complete combustion at relatively low temperatures and excess air levels(25~50%). Second, the residence time of the large lumps of sewage sludge in the hot bed is long enough to ensure sufficient burn-out of the sludge. Third, the freeboard, which acts like a post-combustion chamber, provides complete thermal destruction of the organic substances. Fourth, FBC has lower maintenance cost than other incinerators units.

Sewage sludges may present significant concentrations of potential toxic metals, depending on their origin and wastewater treatment technology. One of the major concerns is the formation of fine inhalable particles enriched with toxic metals. At the high combustion temperature in the incinerating plants, most of the heavy metal compounds are vaporized, but later they condense on the surface of the ash particles in the cooler part of the steam evaporator and are removed with ash (Hirth et al., 1990). Analysis has shown that about 78~98% of Cd, Cr, Cu, Ni, Pb and Zn present in the sewage sludge are retained in the ash, whereas up to 98% of mercury (Hg) may be released into the atmosphere with the flue gas (Forstner et al., 1985, Kozinki et al., 1995). Hg is vaporized to elemental Hg inside the furnaces, and as the flue gas cools, while passing over heat exchanger and flue gas treatment processes along the furnace, oxidation may take place generating Hg^{+} or Hg^{2+} species which may give e.g. HgO, $HgCl_2$ or Hg_2Cl_2 and $HgSO_4$ (Saenger et al., 1999) depending on the availability of Hg, S and Cl and temperature. Hg may vary between 0.1 and 89 mg/kg, Pb from less than 80 to 26000mg/kg and Cd may reach 1100 mg/kg (Dewling et al., 1980).

In this paper, the combustion characteristics of sewage sludges by emission measurements were investigated in commercial FBC units with the capacity ranging from 50 to 160 tons per day. Especially, the emission characteristics of heavy metals including Hg from sewage sludge combustion in a typical FBC plant burning industrial and municipal sewage sludge was investigated by measuring emission concentrations of HAPs at two sampling points which were located at inlet of air pollution control device (APCD) and stack, to observe the performance of an APCD system.

EXPERIMENTAL

1. Investigated Facilities

The incineration conditions of FBC plants treating sewage sludge from different cities were investigated, as presented in Table 1. FBC plants have the capacity ranging from 64 ton to 160 ton per day and the temperature of incineration was around 900℃. Plant 1 and Plant 4 were treating municipal sewage sludge, whereas Plant 2 and Plant 3 were treating the mixed sludges from municipal water treatment in a city and industrial waste water facilities in industrial complex. The moisture content in sludges before dewatering and drying process ranged from 80% to 84%. Table 2 presents the proximate analysis and low heat value (HLV) of sewage sludge which treated in FBC Plants. Carbon content (%) in sludge ranged from 32.1 to 44.5, and the heat value (kcal/kg) ranged from 168 to 239.7. HAPs emission was sampled in FBC plant 2, which treated municipal sewage sludge and industrial sewage sludge at the same time.

2. Sampling and Analysis

2.1. Incineration Process

Fig.1 shows the sewage sludge FBC incineration process with sampling points. Sludge waste after dewatering and drying process was injected inside incineration furnace around the temperature of 900℃. Heat recovery boiler is consisting of steam drum, tube tank, and circulation pump. Superheat combustion gas passed around the tube tank of boiler water, and generated hot steam by indirect heat exchange. The generated steam was recycled for drying process of sewage sludge to increase heat recovery efficiency. The ash particles were generated by the reaction between SO_2 and HCl gas with lime slurry injected in flue gas after passing heat recovery boiler. The most ash particles were captured in Bag Filter (B/F); and SO_2 and HCl in flue gas were captured in wet scrubber. As shown in Fig 1, HAPs sampling points were located at the before B/F and the stack.

Table 1 Sludge Incineration Conditions of FBC Facilities in Operation

Facility	Plant 1	Plant 2*	Plant 3	Plant 4
Capacity(t/d)	64	160	100	85
Temperature(℃)	900	939	923	882
Sludge Injection	S	S+I	S+I	S
Sludge Moisture	80	81	84	83
Daily Ash(kg/d)	6,400[a]	6,093[b]	9,140[b]	6,465[a]
APCD Configuration	SD+BF+WS	SD+BF+WS	SD+BF+WS	C+SD+BF

a : bottom ash + fly ash, b : only bottom ash, S : Municipal Sewage Sludge, I : Industrial Sewage Sludge
C : Cyclone, SD : Semi Dry Reactor, BF : Bag Filter, WS : Wet Scrubber.
* : HAPs Sampling Plant

Table 2 Characteristics of Sewage Sludge Wastes in FBC Facilities

Wastes	C(%)	H(%)	O(%)	N(%)	S(%)	Cl(ppm)	HLV(Kcal/kg)
Sludge 1	32.1	5	19.1	5	0.8	931	168
Sludge 2*	36.7	5.6	18.7	5.1	2.8	2304	235
Sludge 3	40.1	6.5	25.2	5.3	0.3	1521	240
Sludge 4	44.5	6.6	21.8	5.9	0.5	833	204

* : Injection sludge to HAPs sampling Plant

2.2. PM and Heavy Metals

The sampling method for TPM used in the experiment was the Korean standard method for Air Pollution (NIER, 1999), which is similar to the US EPA method 5(US EPA 1995; US EPA Method, 2000). The Korean method uses the thimble-type filter; while the US EPA Method 5 uses the circle-type filter. Dust was collected using an iso-kinetic sampling method, and the factor (%) was ranged from 95 to 110. A PM 2.5 Cyclone (Apex Instrument Co. Ltd.) was used for PM 2.5 sampling, from which the particle size distribution (PSD) was determined. It was adapted for EPA method 201A-Determination of PM2.5 emission and size distribution (US EPA Method 1997). The emission sampling at each point was repeated more than twice. The chemical balance ranged 0.1mg was used to weigh the particle sample from the TPM and PM 2.5 sampling. Heavy metals in particulates were analyzed by ICP/MS(Varian Co.

Ltd., Ultra mass 700) after pre-treatment of samples as documented in EPA method 3050B(US EPA Method, 1986).

2.3. Mercury

Gaseous Hg sampling was conducted in accordance with the OH method (ASTM D 6784). The Hg emission was sampled at two points which was before B/F and Stack. Hg speciation was implemented by OH method at each sampling points. Liquid and solid samples were collected at each process location from the FBC plant 2 and were treated by US EPA 7470A and US EPA 7471. Cold vapor atomic absorption (CVAA) type Lab 254 mercury analyzer (mercury instrument GmbH, Germany) was employed to analyze the total Hg concentration in liquid and solid samples. The sampling point and location for each sample is indicated in Fig 1.

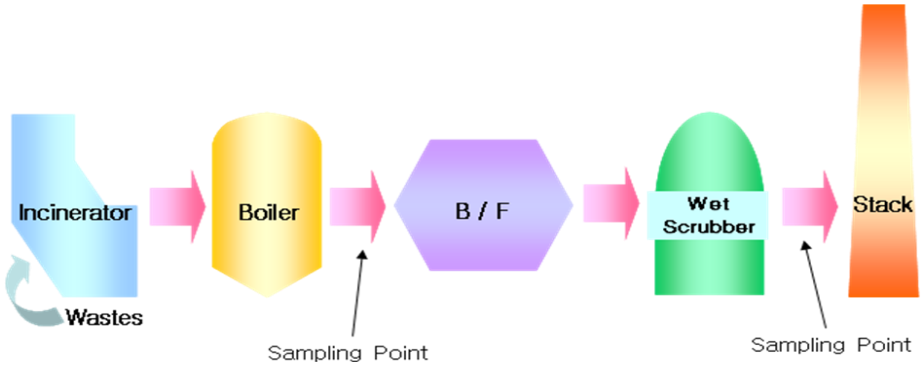

Fig 1. Sludge Incineration Process and HAPs Sampling Points in FBC Plant 2

RESULTS AND DISCUSSION

Table 3 presents the gaseous concentration in flue gas, measured at the stack TMS of FBC plants. For the plants burning industrial sewage sludge, it seemed to emit more NOx and HCl due to higher content of Cl in feedstock. However dioxin concentrations were not negligible due to proper combustion and higher removal efficiencies of APCD for dioxin. Other primary air pollutants such as SOx and CO also very low at stack, which were enough to meet emission regulatory limit. For particulate matter including fine particles will be discussed later, higher concentration of particulate matter was shown in FBC plants when comparing other types of incinerators. Similarly particulate matter is well controlled by passing APCD. So the combustion of sludge in FBC plants was performed well with appropriate gas cleaning system.

Table 4 shows the emission measurements of PM and metals pollutants at inlet of APCD and stack in Plant 2, to observe removal efficiencies of them. Not shown in table 4, in case of NOx, the concentrations at inlet and outlet were 73.3 ppm and 39.5 ppm, respectively. The control efficiency of APCD was 45% in average. SOx concentration at the inlet was 2102ppm, and the outlet was non-detected, which means the control efficiency of APCD was close to 100%. The O_2 concentration at stack ranged 7.3~9.0%, and the CO concentration ranged 24.0~46.0%. Table 4 presents the concentration of PM and heavy metals at each emission sampling point. In case of heavy metals, As, Cd, Cr, Ni, Pb compliance with Korean HAPs emission limit were analyzed. Before APCD, the distribution of heavy metals in TPM and PM2.5 was Cr>Ni>Pb>As>Cd. In case of Cr, the concentration in TPM and PM2.5 were 134mg/Sm^3

Table 3 TMS Data of Operating Facilities in Stack

Plant No.	H_2O (%)	O_2 (%)	CO (ppm)	SOx (ppm)	NOx (ppm)	HCl (ppm)	Dioxin (ngTEQ/Sm^3)
Plant 1	16.48	10.77	16.94	1.01	9.97	0.97	0.01
Plant 2	23.00	11.16	30.66	1.59	45.59	3.94	0.01
Plant 3	13.00	11.86	23.22	0.53	19.91	1.29	0.00
Plant 4	27.82	10.79	0.57	2.60	11.69	2.45	0.00

Table 4 Concentrations at Each Sampling Point and HAPs Control Efficiency

	PM(mg/Sm^3)		AS(μg/Sm^3)		Cd(μg/Sm^3)	
	TPM	PM2.5	TPM	PM2.5	TPM	PM2.5
Before B/F	12371.4	2564.6	110.0	72.6	81.3	38.4
Stack	1.3	1.0	0.9	N.D.	N.D.	N.D.
Efficiency(%)	100.0	100.0	99.2	100.0	100.0	100.0
	Cr(μg/Sm^3)		Ni(μg/Sm^3)		Pb(μg/Sm^3)	
	TPM	PM2.5	TPM	PM2.5	TPM	PM2.5
Before B/F	134054.8	50710.5	19872.9	8495.3	4702.0	1903.9
Stack	5.6	5.4	2.3	N.D.	7.6	N.D.
Efficiency(%)	100.0	100.0	100.0	100.0	99.8	100.0

and 51mg/Sm^3 which means the most enriched metals in TPM and PM2.5, respectively. Ni and Pb were retained in the TPM and PM2.5. At the high combustion temperature in the FBC plants, most of heavy metal compounds are vaporized, and condensed on the surface of the ash particles. About 78~98% of Cr, Ni, Pb, Cd in sewage sludge are vaporized and retained in the ash (Fönster et al., 1985). The control efficiency of APCD investigated was more than 99% for capturing PM and heavy metals.

Fig 2 shows the distribution of PM and heavy metals at the sampling point before APCD. More than 80% of PM was composed of particles larger than 2.5μm. It might be due to the reason that lime was injected in the downstream, after boiler, for capturing SO_2 and HCl and by which PM was generated by the reaction with lime and acid gases. As shown in Table 4, the concentration of heavy metals in PM2.5 was twice more than in TPM. This indicates that the one of the major pathway for generating fine particulates is by the mechanism of the vaporization and condensation of heavy metals. At the outlet of APCD, the PM less than 2.5μm was contributing most of the TPM.

Table 5 presents the concentration and the speciation of Hg at the inlet and outlet of APCD. Sewage sludge normally contains 1-4mg of Hg per kg (dry) which exists in various compounds (Fahlke et al., 1995). As shown in Table 5, oxidized Hg in an average was 93.5% (at inlet APCD) and 6.9% (at stack emission). It is considered that Hg vaporized in the FBC combustion zone. At downstream after boiler, the conversion to oxidized Hg might occur. Due to their low boiling temperature, in the combustion furnace, Hg compounds readily vaporize and exist in gas form after combustion. However, due to the instability of Hg compounds in the gaseous form at higher temperature, more often at above 700℃ the compounds decompose to form elementary Hg (Galbreath et al., 1996, Shaub et al., 1993, Hall et al., 1996). Elemental Hg is not readily soluble and unlike other heavy metals, is not removed with the ash during

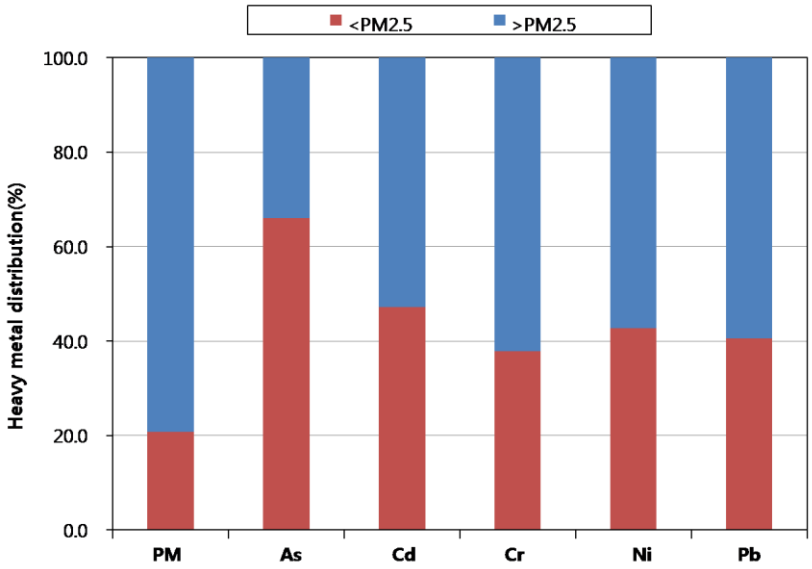

Fig 2. Particulate and Heavy Metals Distribution in Different Sizes before APCD

Table 5 Mercury Concentration and Speciation by OH Method

Sampling Point	Sampling Method	Hg Conc. ($\mu g/Sm^3$)	Hg Distribution($\mu g/Sm^3$)			Hg Speciation(%)		
			Hg^p	Hg^0	Hg^{2+}	Hg^p	Hg^0	Hg^{2+}
Before B/F	OHM-1	1714.2	82.8	31.9	1599.5	4.8	1.9	93.3
	OHM-2	1946.9	39.4	85.7	1821.8	2.0	4.4	93.6
	Average	1830.6	61.1	58.8	1710.7	3.3	3.2	93.5
Stack	OHM-1	1.6	0.03	1.5	0.1	1.8	91	7.2
	OHM-2	2.0	0.02	1.9	0.1	1.0	92.3	6.7
	Average	1.8	0.03	1.7	0.1	1.4	91.7	6.9

post-combustion flue gas treatment. However, at downstream as the flue gas cools down, it is possible that elemental Hg reacts with other components such as HCl, SO_2, H_2O and fly ash, finally forming an oxidized form of Hg(Hg^{2+}) such as $HgCl_2$, HgO, etc. (Meij et al., 2002, Lee et al., 2004, Laudal et al.,
2000a,b) Especially, it is considered that Cl compound in sewage sludge contribute converting elemental Hg to oxidized Hg. At the outlet of APCD, particulate Hg speciation ranged from 2.0 to 4.8%. It is considered that oxidized Hg such as $HgCl_2$, $HgSO_4$, and HgO react with fly ash and has tendency to attach in fine particulate. Generally, in coal power plant, particulate Hg can be removed in ESP and lime slurry in wet FGD scrubber. However, FBC plant was operating wet scrubber to remove acid gas by NaOH injection. It might be the reason that speciation of particulate Hg was higher than gaseous Hg. Since oxidized Hg is soluble in water, most of it was removed in wet APCD, leaving behind elemental Hg as dominant (91.7%) in stack emission (Fig 3).

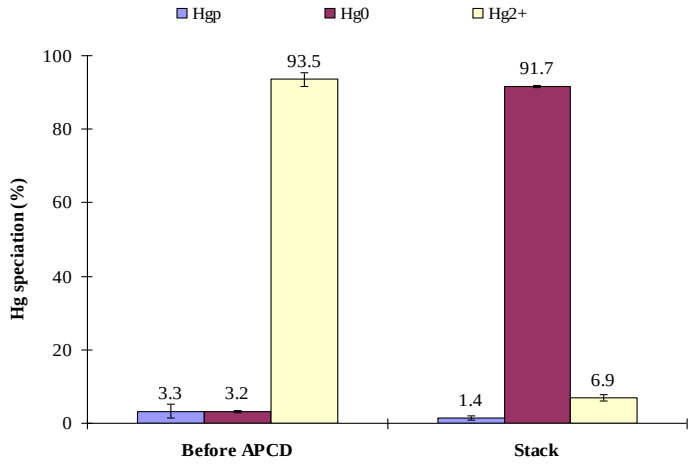

Fig 3 Mercury Speciation Measured by OH Method at Each Sampling Point

CONCLUSION

In this paper, four operating commercial FBC plants were investigated. Among those plants, one typical plant dealing with 150 ton per day was chosen, and combustion characteristics with the emission test of gaseous pollutants such as SOx, NOx, and CO were investigated. Also, the emission of PM, heavy metals and Hg at the inlet and outlet of APCD were sampled and analyzed, respectively. According to the collected data in these tests, the conclusion can be summarized as below.

1. Most of FBC incineration plants burning sludge showed good performances with efficient APCD configuration. In case of burning industrial sludge, NOx and HCl seemed to emit more. However emission of primary air pollutants and HAPs in all plants was low enough to meet regulation limits even though generating higher amount of PM than other types of incinerators.

2. According to the concentration data of TPM and PM2.5 at the inlet and outlet, more than 80% of PM was composed of particles larger than 2.5 μm. It is considered that at the inlet of APCD, coarse particulate matter was generated by the reaction with the acid gas such as SO_2 and HCl and lime powder in the downstream. However, at the outlet of APCD, PM2.5 contributed most of TPM. It is considered that fine particles generated from heavy metals were emitted without being removed by the APCD.

3. The enrichment of heavy metals in TPM and PM 2.5 were Cr>Ni>Pb>Cd>As. The concentration of Cr, which was most enriched heavy metals, was 134mg/Sm^3 and 51mg/Sm^3. The results were related to the original composition of sewage sludge, and the additional tests are required.

4. In case of Hg, more than 93% were speciated into oxidized Hg at the inlet of APCD, and elemental Hg was highest at the outlet of APCD. It is considered that elemental Hg vaporized in combustion zone was converted to oxidized Hg downstream and reacted with flue gases. Oxidized Hg was removed in wet APCD and elemental Hg was dominant in stack emission.

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