

BEHAVIOR OF TRACE METALS IN FLUIDIZED-BED INCINERATION OF SEWAGE SLUDGE

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The behavior of selected heavy metals in sewage sludge during incineration was investigated in a two-stage swirl-flow fluidized-bed incinerator. Leaching tests were also carried out to evaluate the leachability of the metals in the ash discharged from the incinerator.

Selected heavy metals were quantitatively retained in the output stream ash due to both the effects of a low incineration temperature and a uniform temperature distribution in the incinerator. Further, it can be found that the metals contained in the ash were highly stable.

Introduction

In recent years there has been increasing concern about the amount and quality of sewage sludge that must be disposed of. Finding an economically and environmentally acceptable method to dispose of sewage sludge is a pressing need. Of the several methods available to treat and dispose of such sludge, incineration is the most effective one available today. However, it has been recognized that emissions from waste incinerators contain trace quantities of heavy metals. Some of the trace metals such as cadmium, mercury, lead, and zinc are easily volatilized at the high incineration temperatures of conventional incinerators and are then emitted with the flue gas, although large amounts of metal vapors are often condensed on the surface of elutriated fly ash particles as the temperature falls.

Recently, several investigators reported that a fluidized-bed incinerator has the potential to capture the trace metals on the bed material^{8, 12}. Further, selected heavy metals other than mercury can be captured as stable oxides in the incinerator. This may be due to its adequately low operating temperature and intimate contact between solid and gas for effective capture of the metals.

The behavior of trace metals in conventional wastes incineration plants has been studied by many investigators^{2, 3, 9, 10, 14, 15}. Compared with conventional incineration, relatively little data is available on the fate of trace elements during fluidized-bed incineration of sewage sludge^{4, 5, 7}. Therefore, more information on the behavior of trace metals are needed for achieving minimum flux of metals into the environment.

The objective of the present study is to determine

the behavior of trace metals during incineration of sewage sludge in a two-stage swirl-flow fluidized-bed incinerator (TSSF-FBI). The leaching characteristics of trace metals are also investigated to evaluate the stability of incineration residues when finally disposed of.

1. Experimental

1.1 Apparatus and operation

The experimental apparatus for this study is shown schematically in Fig. 1. It has been described in detail previously¹¹. The incinerator was fabricated from stainless steel. The lower and upper stages were divided by an upper-stage distributor. The specifications were: 10.8 cm ID and height of 70 cm for lower stage; 15.8 cm ID and height of 100 cm for upper stage.

For the incineration experiments, the lower bed was filled with sludge-derived ash as the bulk bed material of the lower stage. Initially the upper-stage bed was empty. To start up the incinerator, fluidizing air from the compressor was divided into primary and secondary flows, preheated by the air preheaters and then injected through the lower and upper air distributors into each bed. At the same time, LPG was supplied until the lower bed reached 500°C. Then the air flow and the feed rate of sewage sludge were gradually increased until the lower bed temperature reached a predetermined value. The sewage sludge was fed into the bed by a screw feeder 5 cm above the lower-stage distributor. The ash from the lower bed was removed through an overflow tube 20 cm above the distributor or through an under-bed drain tube installed at the center of the distributor.

Ash from the upper bed was removed through an overflow tube 10 cm above the upper-stage distributor.

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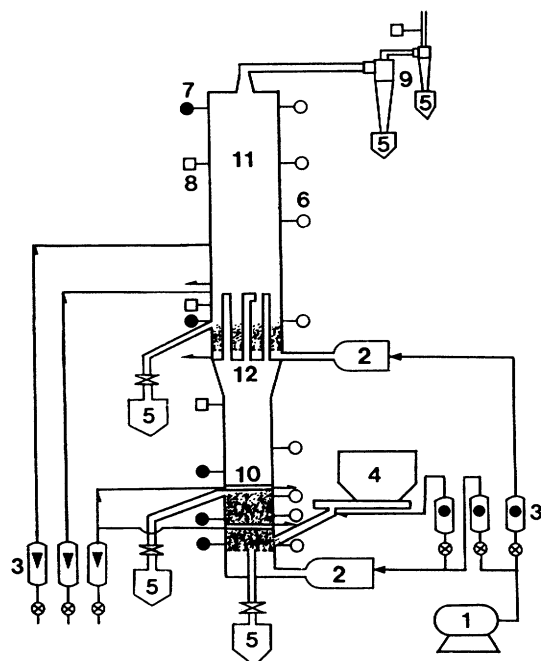


Fig. 1 Schematic flow diagram of TSSF-FBI:
1. air compressor; 2. air preheater; 3. rotameter;
4. sludge hopper and screw feeder; 5. ash hopper;
6. thermocouple and recorder; 7. differential pressure gauge;
8. gas analyzer; 9. cyclones; 10. in-bed cooling line;
11. TSSF-FBI; 12. upper-stage distributor.

The fluidized-bed heights of the lower and upper stages were maintained at 20 cm and 10 cm respectively. The bed temperatures of the lower and upper stages were manually controlled by varying the water flow rate through each cooling line.

Once steady-state incineration conditions were established, the ash drained from the lower and upper stages and collected by cyclones was weighed once an hour and an analysis for heavy metals was carried out. For analysis of metal concentrations in the flue gas, the gas was also sampled.

1.2 Analysis of heavy metals

For analysis of heavy metals in the sewage sludge and the ash from various outflow streams of the incinerator, samples were digested by HF-aqua regia and then analyzed by atomic absorption spectrometry (AAS) or by inductively coupled argon plasma spectrometry (ICP) for heavy metals. Source Assessment Sampling System (SASS) with 0.5M nitric acid solution was used for collecting the gaseous heavy metal elements in the flue gas⁹⁾. Heavy metals were also analyzed by AAS or ICP.

1.3 Leaching test

ASTM D3987-81 batch leaching method¹⁾ was used in this study. This method uses distilled water as the leaching agent. A volume of 40ml of double-distilled water was added to 10g of dry ash to produce a solution with a solid ratio of 4 to 1. The solution was agitated on a mechanical platform shaker to produce constant movement of the mixture. The leaching flask was sealed and closed to the atmosphere for the duration of the 48-hour test. The aqueous phase was separated from the solid

Table 1. Trace metal constituents in sewage sludges and soil ($\mu\text{g/g}$)

Element	Sewage sludge			Soil
	Present study	Ref. 6	Ref. 7	Ref. 16
Cr	207.86	56~14000	130 \pm 10	25
Fe	3.0 $\times 10^4$	(0.88~8.29) $\times 10^4$	(4.1 \pm 0.9) $\times 10^4$	1.3 $\times 10^4$
Cu	620.56	62~2890	500 \pm 100	11
Pb	493	16~3605	430 \pm 20	5.3
Zn	1229.30	71~6890	960 \pm 90	22

Table 2. Concentrations of selected heavy metals in output streams

Element	T_{bl} ($^{\circ}\text{C}$)	C_{x1} ($\mu\text{g/g}$)	C_{x2} ($\mu\text{g/g}$)	C_{x3} ($\mu\text{g/g}$)	C_{x8} ($\mu\text{g/g}$)
Cr	750	426.42	348.45	352.32	ND
	800	388.59	343.44	379.32	ND
	850	406.08	333.38	409.82	ND
Fe	750	42263.90	42254.06	45558.90	ND
	800	45700.44	45720.50	46015.08	ND
	850	46268.90	46016.82	46658.42	ND
Cu	750	932.17	873.96	931.74	ND
	800	894.06	814.28	936.69	ND
	850	900.51	804.20	949.84	ND
Pb	750	799.29	732.58	675.77	ND
	800	765.05	751.96	648.79	ND
	850	723.30	681.58	748.87	ND
Zn	750	1716.42	1651.17	1659.25	ND
	800	1712.68	1674.60	1693.09	ND
	850	1699.25	1700.36	1706.53	ND

ND: not detected.

phase by filtration through a 0.45 μm membrane filter. The leachate was acidified with 0.2ml of concentrated nitric acid and diluted to 100ml with double-distilled water. NaHCO_3 solution (4.0mg/l as HCO_3^-) adjusted with sulfuric acid to pH = 5.7 was also used as a leaching agent to assess the leachability by rainfall when disposed of. This leaching method was the same as the ASTM D 3987-81 procedure.

2. Results and Discussion

2.1 Concentrations of heavy metals in input and output streams

The concentrations of heavy metals in the sewage sludge are listed in **Table 1** and compared with the previous data^{6, 7, 16)}. These studies reveal that concentrations of heavy metals in the sludges are much higher than those in soils. The presence of heavy metals in sewage sludge naturally leads to the question of their origin. It seems that heavy metals in wastewater flows are typically fairly high considering the vast quantities of flow. The heavy metals in the sludge may in part originate from land runoff, industrial wastewater, and domestic sewage.

To determine the behavior of trace metals in sewage sludge during fluidized-bed incineration, it is

Table 3. Mass balances of heavy metals for input and output streams ($T_{b1} = 800^{\circ}\text{C}$, $T_{b2} = 800^{\circ}\text{C}$, $\lambda_T = 1.2$, $U_{o1} = 1.2\text{m/s}$)

Element	Input ($\mu\text{g}/\text{min}$)	Output ($\mu\text{g}/\text{min}$)				Total recovery (%)
	X_s	X_1	X_2	X_3		
Cr	28.75	21.86	7.87	4.55		119.23
Fe	4118.88	2570.65	1047.46	552.18		101.25
Cu	85.79	50.29	18.66	11.24		93.47
Pb	68.13	43.03	17.23	7.79		99.88
Zn	169.95	96.34	38.37	20.32		91.21

necessary to analyze the concentrations of heavy metals in the various output streams. In TSSF-FBI, the output streams include overflow ash from the lower and upper stages, cyclone catch ash, and flue gas.

Table 2 shows the concentrations of heavy metals in the various output streams from the incinerator. In previous results measured in conventional incinerators^{9, 13}), the concentrations of volatile elements such as Cd, As, Cu, Pb, Hg, and Zn were generally found to be higher in those ash discharge streams consisting of finer particles (for example bag filter ash, as compared with bottom ash). This is because the volatile compounds of Cu, Pb, and Zn or the elemental metals themselves are partly evaporated in the incineration zone and then condensed onto the fine ash emitted with the flue gas when the temperature falls. Compared with the behavior of trace metals in conventional incinerators, in the TSSF-FBI the concentrations of selected heavy metals in the ash drained from the lower and upper stages and collected by cyclones were almost equal, as shown in Table 2. This result may be caused by the effects of both a low incineration temperature ($750\sim 850^{\circ}\text{C}$) and a uniform temperature distribution in the TSSF-FBI. Moreover, no metal content was detected in the flue gas. With the observed results, it is noted that the heavy metals such as Cr, Fe, Cu, Pb, and Zn, can be quantitatively retained by the output stream ash during incineration of sewage sludge in the TSSF-FBI.

2.2 Partitions of heavy metals

To determine the elemental balances, the mass flow rates of the various streams, i.e., sewage sludge fed to the incinerator, ash discharged from the lower and the upper stages, cyclone catch ash, and flue gas, were measured in a TSSF-FBI. Samples taken from these output streams were analyzed for selected heavy metals. Since the amount of emitted fly ash which was not collected by cyclones was negligibly small and no metals content in the flue gas was detected, in this study the amount of metals in the emitted fly ash and flue gas was omitted in the calculation of mass balance.

Table 3 shows the material balances for selected heavy metals. As shown in the table, the material balances for Fe and Pb show 100% recovery of metals entering the incinerator. Recoveries of Cu and Zn were somewhat lower than for other metals. This result seems to be due to neglect of the amount of metals in the emitted fly ash which is not collected by cyclones and of

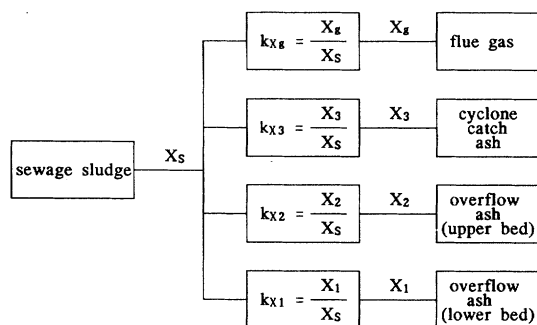


Fig. 2 Partition of an element (X) during incineration of sewage sludge in a TSSF-FBI: sewage sludge(s), overflow ash of lower stage (1), overflow ash of upper stage (2), cyclone catch ash (3), and flue gas (g); $k_{X1} + k_{X2} + k_{X3} + k_{Xg} = 1$.

Table 4. Partition coefficients of heavy metals ($T_{b1}=800^{\circ}\text{C}$, $T_{b2} = 800^{\circ}\text{C}$, $\lambda_T = 1.2$, $U_{o1} = 1.2\text{m/s}$)

Element	k_{X1}	k_{X2}	k_{X3}	$1 - \sum k_{Xi}$
Cr	0.76	0.27	0.16	-0.19
Fe	0.62	0.25	0.13	0.00
Cu	0.59	0.22	0.13	0.06
Pb	0.63	0.25	0.11	0.01
Zn	0.57	0.23	0.12	0.08

$$\sum k_{Xi} = k_{X1} + k_{X2} + k_{X3}$$

metals condensed onto the inner duct surface in the calculations of material balance. On the contrary, over 100% recovery of Cr was obtained. This seems to be due to the surface erosion of stainless steel parts during incineration. A number of published data show that some excess of Cr and Ni was present in the fly ash when the incinerator was made of stainless steel⁹).

Total recoveries of metals in the present study were found to be higher than those reported by other investigators^{5, 13}). With the observed high recoveries, it is noted that two-stage swirl-flow fluidized-bed incineration has potential for reducing trace metal emissions due to less volatilization occurring at a low incineration temperature.

During incineration of sewage sludge in a TSSF-FBI, any element in the sludge can be partitioned into the various output streams as shown in **Fig. 2**. The partition coefficient k_{Xi} is defined as the mass flow ratio of an element in any output stream to that of the sludge fed.

The data in Table 2 were used to calculate the partition coefficients for selected heavy metals. The results are presented in **Table 4**, which shows that selected heavy metals are mostly retained by the ash discharged from the lower and upper stages during incineration of sewage sludge in a TSSF-FBI.

2.3 Leaching characteristics

For evaluating the leachability of heavy metal elements in the ash residues discharged from incineration of sewage sludge, batch leaching tests for the various stream ash and sewage sludge samples were carried out. The batch leaching method used in the present study is a rapid laboratory procedure that is used to roughly esti-

Table 5. Mean concentrations of selected heavy metals in leachates (mg/L)

Leaching solution	Element	Sewage sludge	Ash			*Drinking water standards
			C_{L1}	C_{L2}	C_{L3}	
Distilled water (pH = 6.9)	Cr	0.109	ND	ND	ND	0.05
	Fe	5.340	0.012	0.005	0.024	0.3
	Cu	1.177	0.025	0.007	0.020	1.0
	Pb	0.355	0.035	0.048	0.061	0.05
	Zn	1.158	0.017	0.016	0.029	5.0
Carbonate solution (pH = 5.7)	Cr	0.186	ND	ND	ND	
	Fe	4.963	0.030	0.018	0.039	
	Cu	2.726	0.028	0.008	0.019	
	Pb	1.011	0.048	0.008	0.032	
	Zn	1.197	0.003	0.003	0.010	

ND: not detected.

*: World Health Organization (WHO) guidelines.

mate the concentrations of water-soluble elements in various wastes that can potentially contaminate groundwaters or soils. After leaching the residues, the resulting leachate is analyzed and compared to Resource Conservation and Recovery Act (RCRA) standards. The RCRA standard, in general, is defined as 100 times the drinking water standard. If the concentrations of heavy metals in leachate is beyond the RCRA standards, the waste is prescribed as hazardous wastes. In the present study, both distilled water and NaHCO_3 solution were used as leaching agents.

Table 5 shows the results of leaching tests for sewage sludge and output stream ash. As shown in Table 5, the concentrations of heavy metals in leachate of the sewage sludge are higher than the drinking water standards for both leaching agents although they do not exceed the RCRA standards. For the leachates of various output stream ash, concentrations of heavy metals for both leaching agents are much lower than those in sewage sludge. From the observed results it is found that two-stage swirl-flow fluidized-bed incineration reduces the leachability of selected heavy metals.

Conclusions

During incineration of sewage sludge in the TSSF-FBI, the heavy metals in the sludge can be quantitatively retained in the output stream ash. This is due to both a low incineration temperature and a uniform temperature distribution in the incinerator. The leachability of heavy metals contained in the ash is much lower than that in sewage sludge. Therefore, it is noted that the two-stage swirl-flow fluidized-bed incineration of sewage sludge is a favorable process.

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Nomenclature

C_{L1} = heavy metal concentrations in leachate for ash drained from lower bed [mg/L]

C_{L2} = heavy metal concentrations in leachate for ash drained from upper bed [mg/L]
 C_{L3} = heavy metal concentrations in leachate for fly ash collected by cyclones [mg/L]
 C_{Xg} = gaseous heavy metal concentrations in flue gas [$\mu\text{g/g}$]
 C_{X1} = heavy metal concentrations in ash drained from lower bed [$\mu\text{g/g}$]
 C_{X2} = heavy metal concentrations in ash drained from upper bed [$\mu\text{g/g}$]
 C_{X3} = heavy metal concentrations in fly ash collected by cyclones [$\mu\text{g/g}$]
 k_{Xg} = partition coefficient of heavy metals in flue gas [—]
 k_{X1} = partition coefficient of heavy metals in ash of lower bed [—]
 k_{X2} = partition coefficient of heavy metals in ash of upper bed [—]
 k_{X3} = partition coefficient of heavy metals in cyclone catch ash [—]
 T_{b1} = bed temperature 20 cm above the lower-stage distributor [$^{\circ}\text{C}$]
 T_{b2} = bed temperature 10 cm above the upper-stage distributor [$^{\circ}\text{C}$]
 U_{o1} = primary superficial gas velocity [m/s]
 X_g = mass flow rates of heavy metals in flue gas [$\mu\text{g/min}$]
 X_S = mass flow rates of heavy metals in sewage sludge [$\mu\text{g/min}$]
 X_1 = mass flow rates of heavy metals in ash drained from lower bed [$\mu\text{g/min}$]
 X_2 = mass flow rates of heavy metals in ash drained from upper bed [$\mu\text{g/min}$]
 X_3 = mass flow rates of heavy metals in fly ash collected by cyclone [$\mu\text{g/min}$]
 λ_T = ratio of total air flow rate to theoretical air flow rate [—]

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