

Metal behavior during vitrification of incinerator ash in a coke bed furnace

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Abstract

In this study, municipal waste incinerator ash was vitrified in a coke bed furnace system and the behavior of metals was investigated. Coke and lime were added to provide heat which facilitated vitrification. Ash contributed more than 90% of metal (except for Ca) input-mass. Metal species with low boiling points accounted for the major fraction of their input-mass adsorbed by air pollution control devices (APCDs) fly ash. Among the remaining metals, those species with light specific weights in this furnace tended to be encapsulated in slag, while heavier species were mainly discharged by ingot. Meanwhile, the leachability of hazardous metals in slag was significantly reduced. The distribution index (DI) was defined and used as an index for distribution of heavy metals in the system. A high DI assures safe slag reuse and implies feasibility of recovering hazardous heavy metals such as Cr, Cu, Fe, Pb and Zn. The vitrification in a coke bed furnace proved to be a useful technology for the final disposal of MSW incinerator ash. The heavy metals are separated into the slag, ingot and fly ash, allowing safe reuse of the slag and possible recovery of the metals contained in the ingot and ash fractions.

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1. Introduction

Incinerator flue gas and ash pose a particular environmental threat of dioxins and hazardous metals release. Stringent regulations have been enforced to control flue gas for decades [1], but proper management of residue was often ignored. In numerous countries, the public are seriously concerned about the problem of dealing properly with ash. Direct landfilling has been identified as an inappropriate solution especially in long-term considerations [2,3]. The melting and vitrification process is considered a proper technology for further stabilization of ash [4].

The melting process can lead to a reduction of ~99% [5] or higher [2,6] of toxicity of dioxins, and also an approximate 10–15% (bottom ash) or 80–85% (fly ash) in volume by its high reaction temperature [7,8]. Although heavy metals are not destructed during thermal treatments such as sintering or vitrification, the mobility of heavy metals in ash

can be drastically reduced [8,9]. Electric melting systems are often a common focus of research and industrial applications. However, a coke bed furnace system with a unique characteristic, reductive and energy-intensive environment in the furnace, has not been extensively investigated.

In our previous work, the mobility and encapsulation of metals during a lab scale vitrification was studied, but the behavior of metal species was not investigated [10]. In this study, metal mass fluxes of input and output material during vitrification of incinerator ash in such a system were estimated. It was hoped that further useful information on metal behaviors for operation might be obtained. In addition, the distribution orientation of metal is also included as an index for the potential of metal recovery.

2. Materials and methods

The capacity of the coke bed furnace system studied was 15 t per day. A detailed process diagram is shown in Fig. 1. Incinerator ash, coke, and lime were stored separately in a temporary reservation tank, mixed at a ratio of 10:2.5:1,

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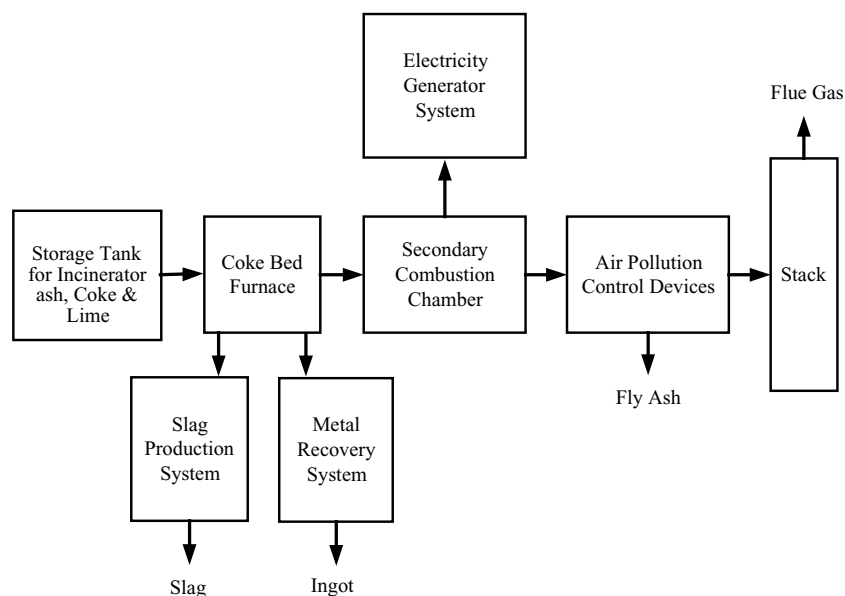


Fig. 1. Process diagram of coke bed furnace.

and then melted at 1600 °C in the furnace. Molten materials, including slag and ingot, were tapped out from different outlets at the bottom of the furnace and soon quenched by water. In this system, 15,000 kg of slag, 1500 kg of ingot, 800 kg of fly ash and 20,000 m³ of flue gas were discharged daily in operation. A secondary combustion chamber and a series of air pollution control devices (APCDs) were used to reduce the emission of air toxic pollutants in flue gas.

The flue gas sampler was equipped with a filter holder, a cooling device, a pump, and a flow meter. The flue gas was sampled isokinetically at an average flow rate of 5 L/min. Details of sampling were given similarly by Lee et al. [11] and our previous work [12,13]. Ambient air was sampled during furnace operation by a standard high-volume sampling train (General Metal Works PS-1) at an average 200 L/min flow rate.

Incinerator ash, coke and lime were directly taken from the temporary storage silo. Slag and ingot were respectively sampled in the slag production and ingot recovery system. Fly ash was collected from each unit of all APCDs. All solid samples were preserved in sealed bottles for further analyses.

Solid samples, including incinerator ash, coke lime, slag, fly ash and ingot, were all pulverized to the size that passed through a 100-mesh sieve (149 µm) to ensure uniform digest efficiency. All samples were all weighed precisely and digested in Teflon vessels hermetically at 180 °C for 5 h with an acid mix, which was composed respectively of 1, 5 and 10 mL of hydrofluoric, nitric and perchloric concentrated acids. After digestion, boric acid was added to destroy the excess hydrofluoric acid. The digests were diluted to exactly 25 mL, filtrated by a 0.8 µm mixed cellulose ester filter and analyzed.

The toxicity characteristic leaching procedure (TCLP) was carried out to assess the mobility of hazardous metal

species in incinerator ash and slag. The extracts of TCLP were digested and diluted to a fixed volume following the detail procedure given in Method 1311 and then analyzed.

Metal species in solid sample digests and TCLP extracts were analyzed with an inductively coupled plasma-atomic emission spectrometry (Jobin Yvon JY-38 Plus ICP-AES). Arsenic and mercury were analyzed by atomic absorption (Perkin Elmer Analyst-100 AA) instead of ICP-AES to achieve lower detection limits. The concentration of elements, including Ag, Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Sb, Se, Si, Sn, Sr, Ti, V and Zn were determined. In this experiment, each sample was analyzed seven times to ensure good reproducibility.

The content of oxygen was analyzed to determine if the slag and ingot were in a non-oxidized state. Relative composition of oxygen and calcium was determined by scanning electron microscopy–energy dispersive spectroscopy (Jeol JXA-840 SEM-EDS). The content of calcium was analyzed by ICP-AES for the approximate mass fraction of oxygen in slag and ingot.

3. Results and discussion

Table 1 shows the metal compositions in incinerator ash, coke, lime and ambient air along with the relative standard deviation (RSD). The compositions of eight hazardous metals, including Ag, As, Cd, Cr, Cu, Hg, Pb and Zn, in ash were 7.32, 597, 70.4, 133, 4280, 3.38, 4570 and 6090 µg/g, respectively. These compositions were roughly similar to those found in other incinerator ash [14], but were 1–2 order higher than those of coke and lime.

Table 2 shows the contribution of each feed material to the mass input of each element. Incinerator ash contributed

Table 1
Compositions of metal species in incinerator ash, coke, lime and ambient air

Element	Mean, $\mu\text{g/g}$ (RSD, %)			Mean, $\mu\text{g}/\text{Nm}^3$ (RSD, %) ^a
	Incinerator ash	Coke	Lime	
Ag	7.32 (13.5)	0.067 (33.3)	0.067 (33.3)	ND ^b
Al	30200 (14.4)	8850 (12.5)	10900 (18.2)	152 (31.6)
As	597 (6.15)	22.8 (15.6)	35.4 (4.47)	12.5 (43.3)
Ba	159 (28.1)	16.0 (13.4)	0.933 (11.2)	5.88 (25.8)
Ca	180000 (27.6)	501 (24.2)	589000 (2.58)	204 (32.3)
Cd	70.4 (13.4)	0.183 (36.8)	0.100 (40.1)	1.93 (47.1)
Co	13.9 (6.92)	0.283 (33.5)	0.117 (35.5)	18.6 (18.8)
Cr	133 (21.4)	24.1 (41.6)	0.350 (14.3)	0.695 (58.1)
Cu	4280 (15.1)	550 (14.5)	258 (34.7)	22.0 (68.4)
Fe	50200 (22.5)	6530 (16.4)	201 (8.30)	302 (39.0)
Hg	3.38 (23.1)	0.356 (44.7)	0.528 (12.5)	0.005 (22.1)
K	10400 (2.05)	1030 (28.5)	4420 (33.6)	44.2 (50.8)
Li	575 (3.27)	24.8 (27.5)	33.6 (10.8)	18.6 (61.3)
Mg	10500 (3.20)	2160 (18.8)	4590 (23.1)	77.0 (74.8)
Mn	658 (9.16)	15.1 (7.79)	32.6 (18.1)	11.9 (39.4)
Mo	26.9 (22.4)	5.68 (34.7)	6.33 (25.8)	ND
Na	10600 (12.5)	1250 (47.5)	2230 (10.2)	1230 (52.5)
Ni	80.6 (22.0)	28.6 (11.2)	15.9 (8.19)	5.59 (36.4)
Pb	4570 (33.5)	1.42 (21.3)	3.88 (25.2)	44.3 (51.3)
Sb	126 (27.1)	2.58 (9.55)	4.36 (33.2)	18.3 (66.2)
Se	33.3 (4.04)	12.1 (32.1)	5.68 (10.8)	4.77 (15.8)
Si	65300 (23.4)	1830 (12.8)	550 (16.7)	892 (18.6)
Sn	99.3 (20.2)	2.36 (44.1)	4.25 (8.56)	12.4 (24.7)
Sr	254 (36.8)	12.8 (12.3)	22.4 (6.36)	1.24 (44.3)
Ti	1020 (12.3)	68.5 (17.2)	134 (35.5)	2.74 (18.4)
V	36.0 (15.0)	6.69 (44.2)	12.4 (11.0)	5.33 (44.2)
Zn	6090 (10.8)	4.40 (10.2)	2.90 (22.6)	66.0 (48.8)

^a RSD: standard deviation/average \times 100%.

^b Non-detectable.

>90% (w/w) of metal species mass, except for Ca. Lime provided 24.7% (w/w) of Ca to facilitate the vitrification and encapsulation. Other metal species mass introduced by coke and lime were not significant. Also, the metal mass contributed by ambient air was found negligible.

Table 3 shows the compositions of metal species in the slag, ingot, fly ash and flue gas. The main compositions in the slag were crust elements, including Ca, Si, Al and Mg, which were 18.9, 10.9, 6.49 and 2.15% (w/w), respectively. The major contents of the ingot were heavy metal species, including Fe and Cu, which were 85.8 and 5.53% (w/w), respectively.

The approximate composition of oxygen in slag and ingot were 10 and 0.2%, respectively. The result clearly pointed out that metal species in slag and ingot were predominately in non-oxidized states, which can be attributed to the reductive capability of coke in this system. The idea of furnace design is referred to an iron-making process.

The compositions of Zn, Ca, Pb and Na in fly ash were 12.1, 9.62, 8.54 and 6.52% (w/w), respectively. The Pb and Zn contents were extremely high in comparison to those in the incinerator ash, implying that recovering by acid or alkaline leaching deserves consideration [15,16]. Both metals can also be reconcentrated to higher levels by recycling the fly ash into the furnace for more economical recovery [10].

The concentrations of hazardous metal in incinerator ash and slag TCLP leachate are shown in Table 4. The leaching concentration of As, Cd, Cr, Cu, Hg, Pb, Se and Zn were 0.794, 1.36, 0.198, 0.806, 0.106, 20.7, 0.027 and 15.8 mg/L in ash, and 0.015, 0.009, 0.015, 0.096, 0.003, 0.026, 0.006 and 0.011 mg/L in slag, respectively. The Cd and Pb leaching levels in incinerator ash exceeded the Taiwanese regulated standards [17]. However, the concentrations of hazardous metal species in slag were relatively much lower. The result showed that immobilization of metals during the vitrification was significant.

The output-mass distributions, distribution tendency and total output-mass balance of metal species are shown in Table 5. In principle, the mass distribution of metals in output-mass was governed by their boiling points and specific weights. Therefore, it is reasonable that metal species with low boiling points, such as As, Cd, Hg, Pb and Zn, were adsorbed by fly ash. Similar results have been found for other furnaces [13]. The fractions of metal mass discharged in flue gas were negligible, indicating high efficiency of APCDs.

Those metals remaining in the furnace would be separated into slag and ingot. The metal species, including Al, Ba, Ca, K, Li, Mg, Mn, Mo, Se, Si, Sr, Ti and V, were mainly tapped out as slag. On the other hand, Cr, Cu, Fe and Ni were mainly discharged along with ingot. The above

Table 2
Mass distribution of each element among the input materials

Element	Mass distribution of input material (wt.%)			
	Incinerator ash	Coke	Lime	Ambient air
Ag	99.7	0.2	0.1	0.0
Al	90.1	6.6	3.3	0.0
As	98.5	0.9	0.6	0.0
Ba	97.5	2.5	0.6	0.0
Ca	75.3	0.1	24.7	0.0
Cd	99.9	0.1	0.0	0.0
Co	99.4	0.5	0.1	0.0
Cr	95.6	4.3	0.0	0.0
Cu	96.3	3.1	0.6	0.0
Fe	96.8	3.2	0.4	0.0
Hg	96.0	2.5	1.5	0.0
K	93.7	2.3	4.0	0.0
Li	98.4	1.1	0.6	0.0
Mg	91.4	4.7	4.0	0.0
Mn	98.9	0.6	0.5	0.0
Mo	92.9	4.9	2.2	0.0
Na	95.2	2.8	2.0	0.0
Ni	90.2	8.0	1.8	0.0
Pb	100	0.0	0.0	0.0
Sb	99.1	0.5	0.3	0.0
Se	90.3	8.2	1.5	0.0
Si	99.2	0.7	0.1	0.0
Sn	99.0	0.6	0.4	0.0
Sr	97.9	1.2	0.9	0.0
Ti	97.1	1.6	1.3	0.0
V	92.5	4.3	3.2	0.0
Zn	100	0.0	0.00	0.0

Table 3
Compositions of metal species of slag, ingot, fly ash and flue gas

Element	Mean, $\mu\text{g/g}$ (RSD, %)			Mean, $\mu\text{g}/\text{Nm}^3$ (RSD, %)
	Slag	Ingot	Fly ash	Flue gas
Ag	2.08 (8.14)	11.5 (55.9)	47.6 (20.9)	1.97 (48.3)
Al	64900 (16.8)	12000 (45.9)	20800 (44.1)	15600 (38.6)
As	40.9 (8.33)	159 (16.6)	4820 (25.8)	569 (24.1)
Ba	146 (21.4)	28.8 (57.3)	68.5 (13.5)	99.1 (51.5)
Ca	189200 (4.43)	20500 (42.7)	96200 (42.5)	1470 (44.6)
Cd	2.53 (26.6)	5.79 (6.05)	1690 (15.7)	66.9 (29.6)
Co	5.72 (7.07)	136 (12.3)	8.76 (69.5)	20.3 (32.5)
Cr	13.3 (29.6)	2420 (8.87)	81.6 (58.3)	93.1 (38.7)
Cu	5.21 (9.05)	55300 (19.6)	4060 (40.1)	1.73 (58.8)
Fe	9630 (41.3)	858000 (1.47)	28500 (23.4)	21800 (21.5)
Hg	0.033 (38.9)	0.125 (29.9)	77.6 (10.6)	0.254 (28.6)
K	3250 (1.53)	194 (40.4)	26800 (31.4)	335 (24.7)
Li	580 (6.09)	113 (23.2)	252 (41.7)	45.6 (39.4)
Mg	21500 (7.91)	328 (56.8)	8550 (63.5)	11.2 (44.5)
Mn	1130 (7.51)	2860 (50.8)	363 (38.7)	627 (28.0)
Mo	29.1 (17.9)	91.4 (11.8)	49.3 (12.4)	41.1 (64.8)
Na	4270 (8.63)	4260 (22.8)	65200 (34.7)	1260 (49.9)
Ni	8.99 (52.7)	1260 (15.2)	178 (52.1)	276 (23.9)
Pb	33.2 (15.6)	182 (11.6)	85400 (18.8)	199 (42.2)
Sb	72.3 (18.3)	192 (6.47)	980 (26.3)	316 (66.1)
Se	38.6 (18.3)	86.3 (13.4)	68.5 (42.9)	22.3 (28.4)
Si	109000 (17.6)	1120 (32.4)	55400 (22.5)	2210 (18.6)
Sn	19.3 (17.6)	854 (22.4)	1810 (31.0)	28.5 (71.2)
Sr	535 (3.88)	11.0 (46.2)	1920 (28.6)	33.4 (5.91)
Ti	1530 (18.1)	1400 (32.4)	663 (45.5)	214 (47.2)
V	38.1 (10.1)	131 (16.1)	67.5 (36.8)	136 (50.1)
Zn	2.55 (31.9)	217 (10.6)	121000 (44.2)	250 (23.9)

Table 4
Concentration of hazardous metal species in the solution of TCLP test

Hazardous metal species	Concentration of hazardous metal species (mg/L)		
	Incinerator ash	Slag	Regulated standard ^a
As	0.794	0.015	5.00
Cd	1.36	0.009	1.00
Cr	0.198	0.015	5.00
Cu	0.806	0.096	–
Hg	0.106	0.003	0.200
Pb	20.7	0.026	5.00
Se	0.027	0.006	1.00
Zn	15.8	0.011	–

^a This is the regulated standard in Taiwan.

results clearly pointed out that light metal species went into the slag, while heavy metal species stayed with the ingot. The separation of metal species can be attributed to gravity [7,18].

In order to describe the distribution and final destinations of hazardous metals during vitrification, the distribution index (DI) is adopted. DI is defined as the maximum value of weight percentage among the slag, ingot, vaporized (fly ash + flue gas) phases. Metals with DI >60% are divided into three categories according to their destinations, namely slag-orientated (SO), ingot-orientated (IO) and vapor-orientated (VO) metal. Those metals with all weight percentages lower than 60% are labeled as non-orientated (NO).

Table 5
Mass distribution of each element among the output materials

Element	Mass distribution of output material (wt.%)					
	Slag	Ingot	Fly ash	Flue gas	Distribution index	Total output-mass balance
Mg	97.5	0.2	2.3	0.0	97.5(SO)	172
Si	97.0	0.1	2.9	0.0	97.0(SO)	154
Al	96.2	2.0	1.8	0.0	96.2(SO)	181
Ca	96.0	1.2	2.9	0.0	96.0(SO)	74.3
Li	95.5	2.1	2.5	0.0	95.5(SO)	93.5
Ba	95.2	2.1	2.6	0.1	95.2(SO)	84.6
Ti	88.7	9.0	2.3	0.0	88.7(SO)	147
Sr	82.3	0.2	17.5	0.0	82.3(SO)	226
Mn	76.9	21.6	1.5	0.1	76.9(SO)	199
Se	73.8	18.4	7.8	0.1	73.8(SO)	127
Mo	68.9	24.1	6.9	0.1	68.9(SO)	131
V	67.0	25.6	7.0	0.4	67.0(SO)	131
K	66.8	0.4	32.8	0.0	66.8(SO)	39.3
Cu	0.1	96.1	3.8	0.0	96.1(IO)	129
Cr	4.6	93.6	1.7	0.0	93.6(IO)	186
Fe	9.0	89.3	1.6	0.0	89.3(IO)	185
Ni	5.6	87.5	6.6	0.3	87.5(IO)	161
Co	26.7	70.7	2.4	0.1	70.7(IO)	137
Zn	0.0	0.3	99.6	0.0	99.6(FO)	106
Hg	0.7	0.3	99.0	0.0	99.0(FO)	119
Pb	0.6	0.4	99.0	0.0	99.0(FO)	138
Cd	2.5	0.6	96.8	0.1	96.9(FO)	132
As	11.8	5.1	82.8	0.2	83.0(FO)	51.2
Ag	33.6	20.7	45.6	0.0	45.6(NO)	75.8
Na	49.6	5.5	44.9	0.0	49.6(NO)	69.8
Sb	47.5	14.0	38.2	0.3	47.5(NO)	108
Sn	8.7	42.9	48.4	0.0	48.4(NO)	198

When a solid waste containing certain hazardous metals at very high concentrations is vitrified, DIs can be used to predict the destinations of these metals. Metal species with high mass DIs tend to be more concentrated in a single phase among slag, ingot, and vapor. In contrast, NO metals tend to be dispersed among various phases in the output material during vitrification.

From the practical point of view, high mass distributions of IO and VO metals imply that these species are potentially recoverable if the concentration levels in the output material are high enough. On the other hand, it also means that the dispersion of hazardous metal is constrained and the possibility of secondary pollutions is reduced.

Since, the hazardous metals in this system were not SO, the slag could be safely reused. The high distribution mass of VO hazardous metals indicated that the efficiency of APCDs is very important from the environmental point of view. The recovery of IO metal species has been done [4], but still disposal of ingots containing Cr at concentration levels should be proceeded with caution.

The total output-mass balance ranged from 39.3 to 226% and averaged 131%. The intrinsic differences between output-mass and input-mass could be probably due to memory effect and sampling error [14]. The sampling error can be explained by the extreme inhomogeneity of ash, which is aggregation of residue. However, the total output-mass

balance may serve as a check for the precision of analysis. While, the mass balance of the metal species was out of the range of three-fold or larger. The analysis of samples and calculation of output-mass balance has to be conformed to ensure the accuracy in the calculation of the metal fates.

4. Conclusion

The main metals in the slag were Ca, Si, Al and Mg, while the major metals in ingot were Fe and Cu. The major metals of fly ash were Zn and Pb, which could lead to their recovery by acid or alkaline leaching. The metal elements existed predominately in non-oxidized states both in slag and ingot. The mobility of hazardous metals in the slag is reduced after vitrification as verified by the TCLP test.

The distributions of metal output-mass in this system are governed by boiling point and specific weight of metal. The fractions of metal mass discharged in flue gas were negligible. During vitrification, metal species with low boiling points, including As, Cd, Hg, Pb and Zn, were mostly adsorbed by the APCDs fly ash. Species including Al, Ba, Ca, K, Li, Mg, Mn, Mo, Se, Sr, Ti and V were mainly tapped out as slag, while the heavy metal species, including Co, Cr, Cu, Fe and Ni, were mainly discharged along with the ingot. The metals were separated between the slag and ingot by gravity.

It is proposed that the distribution index be used to assess the feasibility of metal recovery and may also serve as an index to insure the security for future application of vitrification of other waste in this system. The vitrification in a coke bed furnace proved to be a useful technology for the final disposal of MSW incinerator ash. The heavy metals are separated into the slag, ingot and fly ash, allowing safe reuse of the slag and possible recovery of the metals contained in the ingot and ash fractions.

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