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Coleaching behavior of lead of incineration ash waste mixtures

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Abstract

In this paper, the authors reported the coleaching behavior of lead of incineration ash waste mixtures, i.e., fabric filter residues [called *fly ash* (FA)] and semidry scrubber residues [called *reaction product* (RP)]. The leaching potential of lead metal of those mixtures was estimated by toxicity characteristic leaching procedure (TCLP) with acidic extractant. And it was found to be under the controls of dissolution rate and interparticulate interactions. The author has developed a linear regression equation to fit the coleaching behavior well, the equation is basing on the chemical compositions of (RP/FA) mixtures. The impact of this study is that the leaching toxicity of mixture could be reduced to be nonhazardous, just only to adjust the mixing ratio of these two ash wastes. And the authors are able to use these ash wastes for wider recycling applications without any intermediate stabilization treatment.

Keywords: Lead; Fly ash; Characterization; Reaction; CaO

1. Introduction

Over 90% of refuse wastes in Taiwan are treated by combustion in 19 large waste resource recycling plant (the municipal solid waste incineration). The total generation rate of combustion residues, such as boiler ash, scrubber ash, precipitator ash and bottom ash, has been predicted to be 2000 ton/day [1]. Heavy metal leach behavior of residues of incineration air pollution controlling devices has been studied for a long time [2–9].

Generally, in Taiwan, the mean of fly ash (FA) is the residue mixture that is collected from dry or semidry scrubbers (the flue gas spray dryer/absorber) and electrostatic precipitators or fabric filters. Since 1995, people started to link the relationship of chemical composition with heavy metal leach behavior between these pure residues [10–17]. As we all know, each isolate residue is also a mixture of fine inorganic compound too, such as Al_2O_3, SiO_2, CaO, etc.

Cement-based solidification/stabilization (S/S) process has been recognized to be the best method to process these hazardous ashes. Eighmy et al. [18] and Rahman et al. [19] have reviewed the present state of this art, including its patent literature and fundamental aspects, respectively.

In Taiwan, the solidification operation is usually to add cement (up to a maximum 25 wt.%) and heavy metal chelator into the toxic ashes to full fit the regulatory required unconfined single axial compressive strength (UCS) and toxicity characteristic leaching procedure (TCLP) test. For most of solidified MSWI residue concrete, an important indicator for the long-time stability assessment is the leaching potential of lead metal. A rare study has concerned on the effect of the alkalinity or salinity of residues on the lead metal leaching from the residue. In this paper, we want to elucidate the relationship of the lead metal coleachability with chemical composition of the mixture of fabric filter residues [called *fly ash* (FA)] and semidry scrubber residues [called *reaction product* (RP)].

2. Experimental

Seven municipal solid waste (MSW) incineration plants with treat-capacity 900–1350 ton/day were selected to

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Table 1
Chemical analysis result of APC residues

Plant	SiO_2 (wt.%)		Al_2O_3 (wt.%)		CaO (wt.%)		CaO/SiO_2		CaO/Al_2O_3	
	FA	RP	FA	RP	FA	RP	FA	RP	FA	RP
1	9.26	32.20	26.33	12.0	55.21	38.45	5.96	1.19	2.10	3.20
2	9.12	20.60	16.15	39.31	50.75	26.71	5.56	1.30	3.14	0.68
3	17.57	19.60	32.30	46.31	42.36	32.20	2.41	1.64	1.35	0.69
4	3.12	36.48	42.82	32.63	41.39	26.25	13.27	0.72	0.97	0.80
5	3.76	21.08	37.48	44.71	45.65	26.14	12.14	1.24	1.22	0.58
6	6.80	24.28	16.40	33.59	55.20	38.28	8.12	1.58	3.37	1.14
7	3.91	36.68	37.93	24.61	42.60	28.16	10.90	0.77	1.12	1.14

collect their air pollution control devices (APCDs) residues. A semidry lime scrubbing system following a fabric filter used to clean the flue gas comprises these APCDs. Residues used in this paper are mainly collected from the fabric filter [also called *fly ash* (FA)] and the scrubber [also called *reaction product* (RP)] in these plants. Ashes were collected separately from the sampling hole of the APCD units and aged for 3 days to stable the hydrophilic property of RP. The aging was to keep these residues within the condition of a temperature range with 20–22 °C and a moisture range with 45–65%. The boiler ash must be sure that there are no mixes with EP ash or RP.

2.1. Toxicity leaching extraction procedure test

Toxicity leaching extraction procedure (TCLP) test followed the USA EPA Method # of 1311: the ash sample is mixed with acetic extractant of a pH value of 2.88. The solid-to-liquid ratio is 1:20, following an agitation extraction with a speed of 30 rpm for 18 h. Lead concentration in the final extracts was determined by flame atomic absorption spectrometer following the US EPA Method # of 7420. TCLP test is to simulate the long-term leaching behavior when there is codisposal of hazardous waste with general solid waste in landfill. Ten volume percent of HNO_3 buffer solution extraction (pH=0.1) represents the total lead amount ($[Pb]_o$) that is acid extractable.

2.2. Chemical composition analyzes of ashes

Wet chemical analysis followed ASTM Method of C114-94.

A Fisher Scientific Accruement pH meter was used to probe the pH value of the solution. A Solar-969 atomic absorption (AA) spectrometer made by Unicam in the United States was used to monitor concentrations of heavy metals. All of the fine chemicals were charged from Alderich and were used directly without purify.

3. Results and discussions

Many literatures [18] have published chemical composition analysis by wet chemical, instrumental analysis on pure scrubber residues (RP) and fabric filter residues (FA). Table 1 has summarized the main chemical composition of residues used in this paper. In present, the authors defined the leaching potential of lead metal to be $[Pb]/[Pb]_o$ (see Experimental section). Table 2 lists the TCLP results of tested ashes. It has an advantage in that we could compare the leachability between different ash waste sources, and without considering their particle size, chemical compositions and others complex factors.

Because the residue is Porlozzanic material, the principal hydration product of the TCLP is calcium silicate hydrate ($CaO–SiO_2–H_2O$) gel or calcium aluminate hydrate ($CaO–Al_2O_3–H_2O$) gel. The dissolution model of $CaO–SiO_2–H_2O$ gel had been proposed by Rahman et al. [19], and they concluded that the dissolution of $CaO–SiO_2–H_2O$ gel strongly dependents on CaO/SiO_2 ratio in the range of $CaO/SiO_2>1$. For the experimental data of FA in Table 1, all have a ratio of Ca/Si over 1. Figs. 1 and 2 show the relation of lead leachability and CaO/Al_2O_3 or CaO/SiO_2 ratio. The "activity" means the ability of free species of $Ca(OH)_2$ or

Table 2
Lead metal leaching data of collected ashes

Method	FA			RP		
Plants	$[Pb]_o$ 10% HNO_3	[Pb] TCLP	$[Pb]/[Pb]_o$	$[Pb]_o$ 10% HNO_3	[Pb] TCLP	$[Pb]/[Pb]_o$
1	115.2	12.76	0.110	27.8	2.587	0.0930
2	83.9	4.83	0.056	11.3	0.1087	0.0096
3	126.7	0.11	8.69E−4	31.1	0.261	0.0084
4	263.5	73.30	0.278	133.9	0.696	0.0052
5	147.6	38.04	0.258	22.4	0.065	0.0029
6	79.1	9.72	0.123	27.0	0.63	0.0233
7	172.8	38.61	0.223	17.0	0.46	0.0271

Unit: ppm.

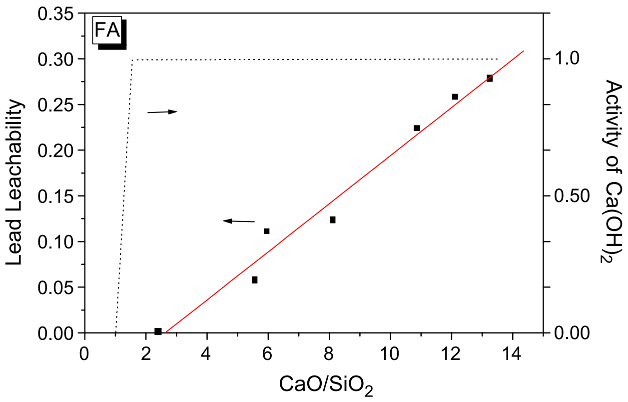

Fig. 1. Plot of CaO/SiO$_2$ ratio versus lead leachability of FA residues.

Al(OH)$_3$ reacting to heavy metals. Two line regressions were obtained from Figs. 1 and 2:

$$\left(\frac{[\mathrm{Pb}]}{[\mathrm{Pb}]_o}\right)_{FA} = -0.0692 + 0.0263 \times \left(\frac{\mathrm{CaO\%}}{\mathrm{SiO_2\%}}\right)_{FA} \quad (1)$$

$$\left(\frac{[\mathrm{Pb}]}{[\mathrm{Pb}]_o}\right)_{RP} = -0.0162 + 0.0342 \times \left(\frac{\mathrm{CaO\%}}{\mathrm{Al_2O_3\%}}\right)_{RP} \quad (2)$$

3.1. Coleaching behavior of binary mixtures

Fig. 3 and Table 3 has shown the leached data of the FA/RP binary mixtures of Plant 7. The [Pb] in this table is slightly different from Table 2, it is caused from different batch experiment for the same sample. The ash wastes are in heterogeneous, so that we can not get the same [Pb] for each batch, but they were in the same order of magnitude. In the system, ratios of CaO/SiO$_2$ and CaO/Al$_2$O$_3$ in the mixtures should be calculated by the following methods:

$$\left(\frac{\mathrm{CaO\%}}{\mathrm{SiO_2\%}}\right)_{\mathrm{mixture}} = \frac{x_{FA} \times (\mathrm{CaO\%})_{FA} + x_{RP} \times (\mathrm{CaO\%})_{RP}}{x_{FA} \times (\mathrm{SiO_2})\%)_{FA} + x_{RP} \times (\mathrm{SiO_2\%})_{RP}}$$

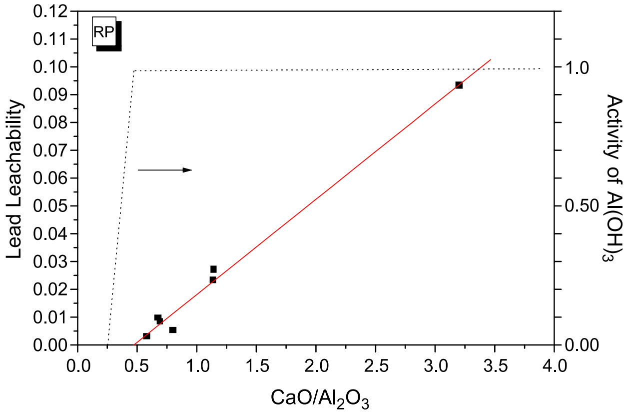

Fig. 2. Plot of CaO/Al$_2$O$_3$ ratio versus lead leachability of RP residues.

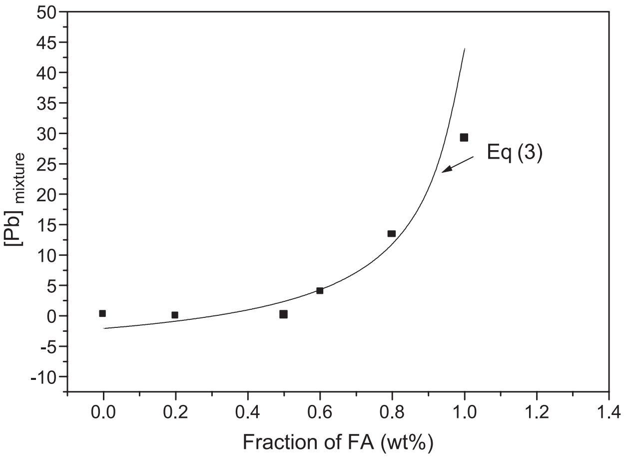

Fig. 3. Plots of coleaching data of binary fly ash/reaction product mixtures from Plant 7 and their theoretical prediction results.

$$\left(\frac{\mathrm{CaO\%}}{\mathrm{Al_2O_3\%}}\right)_{\mathrm{mixture}} = \frac{x_{FA} \times (\mathrm{CaO\%})_{FA} + x_{RP} \times (\mathrm{CaO\%})_{RP}}{x_{FA} \times (\mathrm{Al_2O_3\%})_{FA} + x_{RP} \times (\mathrm{Al_2O_3\%})_{RP}}$$

$$x_{FA} + x_{RP} = 1.0$$

As shown in Fig. 3, the leaching data of lead metal of mixture could be described well by Eq. (3).

$$\begin{aligned}([\mathrm{Pb}])_{\mathrm{mixture}} = {} & ([\mathrm{Pb}]_0)^*_{FA}\left[-\frac{1}{2}\left\{0.0692 \right.\right.\\ & \left.\left. + 0.0263 \times \left(\frac{\mathrm{CaO\%}}{\mathrm{SiO_2\%}}\right)_{\mathrm{mixture}}\right\}\right] \\ & + ([\mathrm{Pb}]_0)^*_{RP}\left\{0.0162 + 0.0342 \right. \\ & \left. \times \left(\frac{\mathrm{CaO\%}}{\mathrm{Al_2O_3\%}}\right)_{\mathrm{mixture}}\right\} \end{aligned} \quad (3)$$

Eqs. (1) and (2) comprised Eq. (3) with a prefactor 0.5 at the first term. It indicates that the lead leaching from isolate particulate and the leaching potential of lead metal of mixed residue is also governed by ratios of CaO/SiO$_2$ and CaO/Al$_2$O$_3$.

Table 3
TCLP data of residue mixture

FA/RP	Plant 7	
	Residual weight (wt.%)	[Pb] (ppm)
100/0	55.48	29.175
90/10	–	–
80/20	57.63	13.4
70/30	–	–
60/40	63.97	3.95
50/50	66.66	0.175
40/60	–	–
30/70	–	–
20/80	81.18	0.079
10/90	–	–
0/100	64.89	0.211

3.2. Coleaching mechanism

In the coal fly ash, the driving force of the metal leaching has been well studied and recognized as it mainly caused the surface dissolution and fast buffering of alkali species to leach the metal ions from particulate surface [20,21]. The authors assume that the metals in the FA and RP follow the same mechanism to leach out. In addition, the equilibrium of leaching in a single particulate was reached. It is difficult to reach equilibrium for the whole sample, due to the environmental and surface structure constraints.

Generally, RP possesses more hydrated lime, $CaSO_4$ and lead concentration. While fly ash possesses high free CaO content or $CaCO_3$ and $Ca(OH)_2$. When RP was mixing with FA, extremely high SiO_2 content of RP (see Table 1) supports extra interaction site to free CaO of FA. And the total leachability of FA will be reduced. Eq. (1) has shown the dependence of the lead leachability and CaO/SiO_2 of FA. Increasing the RP content in FA to increase the reduction of lead leachability in FA, the decay curve in Fig. 3 could be explained.

The impact of this relation is that the leaching toxicity of mixture could be reduced to be nonhazardous, just only to adjust the mixing ratio of these two ash wastes. We are able to use these ash wastes for wider recycling applications without any intermediate stabilization treatment.

4. Conclusions

The leaching potential of lead metal from surface of fabric filter residues [called *fly ash* (FA)] is controlled by compositional ratios of CaO/SiO_2, while the leaching potential of lead metal from surface of scrubber residues [called *reaction product* (RP)] is controlled by compositional ratios of CaO/Al_2O_3, as assessed by TCLP. The linear coleaching behavior of lead metal of FA/RP binary mixture can be described well by our empirical equation, which are developed based on the main chemical composition control mechanism. In conclusion, it is possible to adjust the release toxicity of heavy metal from APC residues by changing the ratio of pure FA/RP.

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