

Evaluation of the distribution patterns of Pb, Cu and Cd from MSWI fly ash during thermal treatment by sequential extraction procedure

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ABSTRACT

Municipal solid waste incinerator (MSWI) fly ash was frequently classified as hazardous materials as the metals' concentration of toxicity characteristic leaching procedure (TCLP) exceeded regulations. Many studies have focused on reducing the concentration of TCLP using thermal treatment and increasing the application of thermally treated slag. However, the metal patterns in MSWI fly ash with or without thermal treatment have seldom been addressed. The main objective of this study was evaluation of the distribution patterns of Pb, Cu and Cd from MSWI fly ash during thermal treatment by sequential extraction procedure. The experimental parameters included the form of pretreatment, the proportion of bottom ash (bottom ash/fly ash, B/F = 0, 0.1 and 1) and the retention time. The results indicated that (1) In comparison to raw fly ash, the distribution patterns of Pb, Cu and Cd become stable in thermally treated slag. (2) Washing pretreatment caused the Pb pattern to become stable, while the influence on Cu and Cd were not significant. (3) The distribution patterns of Pb, Cu, and Cd became more stable as the retention time increased. (4) Adding bottom ash could make the distribution patterns of Pb and Cd more stable.

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1. Introduction

Because of the recent economic development in Taiwan, a great deal of garbage needs to be appropriately treated and disposed. As Taiwan is a small island country having a high population density, solid waste is primarily dealt with by incineration. The Taiwan government has established approximately 22 municipal solid waste incinerators (MSWI). Incineration has many advantages, such as reducing waste volume and detoxifying waste. However, incineration also generates some secondary pollutants such as gases and particle pollutants. The gases and particle pollutants generated are NO_x and SO_x, and fly ash and bottom ash, respectively. During the combustion process, some metal compounds are vaporized at high temperature and condensed on MSWI fly ash at low temperature in an air pollutant control device. The metals' concentration in toxicity characteristic leaching procedure (TCLP) test leachates of MSWI fly ash, especially for lead, often exceeds the Taiwan EPA regulation. Therefore, MSWI fly ash is considered a hazardous material [1–3]. As a great quantity of MSWI fly ash is produced annually, numerous studies have investigated MSWI fly ash treatments to reduce the metals' concentration in the TCLP test leachates and increase

recycling and the reusing potential of MSWI fly ash. Some studies evaluated pollutant emissions during the treatment process [4–10]. In addition to treatment methods for MSWI fly ash, the leachability of heavy metals from treated slag is also a great concern. The sequential extraction procedure developed by Tessier et al. [11] is often used to identify the speciation of heavy metals. This method focuses on the breakdown of materials into varied fractions that can be selectively dissolved by using particular extraction agents. The fractions are often defined as follows:

- (I) The exchangeable fraction is easily extracted with solutions in neutral condition and changes in water ionic composition are likely to affect the sorption–desorption processes.
- (II) The carbonate fraction is associated with sediment carbonates. This fraction is soluble under acidic conditions and susceptible to changes of pH.
- (III) The iron and manganese oxides are excellent scavengers for trace metals and are thermodynamically unstable under anoxic conditions. This fraction can extract iron with a reducing solution followed by an acid or a complex agent.
- (IV) Trace metals may be bound to various forms of the organic matter fraction (notable humic and fulvic acids). In order to release these metals, the organic matter can be degraded under oxidizing conditions. Hydrogen peroxide is generally used as extraction agent for such organic matter.

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(V) The residue contains mainly primary and secondary minerals that retain metals in their crystalline structure. These metals in residue are not normally expected to be released into the environment under natural conditions and can be decomposed by digestion with oxidizing acids.

Sequential extraction procedure was applied to solids, sludge, MSWI fly ash and bottom ash. Fuentes et al. [12] applied sequential extraction to different sewage sludges obtained from wastewater treatment plants. Analytical results by Fuentes showed that chromium and copper were most abundant in organic and residual fractions, while zinc and nickel had no dominant chemical phase. Dermatas et al. [13] reported that the percentage of the Pb fraction in soil followed the sequence: carbonate fraction > iron and manganese oxides fraction > organic matter > exchangeable > residual. Over 95% of extracted Pb was associated with carbonate, iron and manganese oxide fractions. Previous studies that investigated the release of metals from MSWI bottom ash determined that almost all Cr and major fractions of Mn were extracted with strong acid. Copper was extracted under oxidizing conditions, and Pb and Zn were acid soluble and easily released into the environment [14,15]. Moreover, some studies have focused on metal fractions in MSWI fly ash using sequential extraction. The experimental results obtained by these showed that most fractions of toxic metals, such as Cu, Cd, Zn, and Pb, accumulated in exchangeable and carbonate fractions. The main fraction of Cd was bound to carbonate and soluble compounds. The chief fractions of Pb, Zn, and Cu were bound to carbonate and organic matter. The physical and chemical properties of MSWI fly ash, such as particle size and composition, also influenced the decomposition of metals obtained by sequential extraction [16–24]. Many studies have focused on metal fractions using the sequential extraction method. However, very few reports have compared metal fractions in raw ash and treated slag. Chou et al. [25] evaluated the emissions of Pb and polycyclic aromatic hydrocarbons (PAHs) from thermally co-treated MSWI fly ash and bottom ash process. In that study, the experimental results only showed the TCLP concentration of Pb. However, the metal fractions in raw fly ash and thermally treated slag with different operational conditions were less to mention.

The aim of this study is to evaluate the patterns distribution of Pb, Cu, and Cd patterns from MSWI fly ash during thermal treatment by sequential extraction procedure. Thermally treated and untreated metal fractions are compared. In addition to thermal treatment, the effects of parameters, such as retention time, pretreatment, and co-treatment on metal fractions are also discussed.

2. Experiment

2.1. Materials

The MSWI fly ash and bottom ash samples used in this study were from a mass-burning incinerator in Taiwan. The MSWI fly ash and bottom ash were sieved to ensure that they contained particles of the same size, respectively. The full chemical and mineralogical characterization of raw fly ash, washed fly ash and bottom ash listed in Table 1 were performed and published in the previous papers [4,26].

2.2. Apparatus

Thermal treatment of the fly ash was conducted in laboratory apparatus and the experiment apparatus of this study was similar to that used by Chou et al. [25]. This apparatus was a pilot-scale rotary kiln reactor with a thermal chamber (210 cm long with an internal

Table 1

Main composition of raw fly ash, washed fly ash and bottom ash

Composition (%)	Raw fly ash	Washed fly ash	Bottom ash
CaO	35.81	41.54	18.21
SiO ₂	7.62	15.15	24.49
Al ₂ O ₃	3.38	7.29	2.67
Fe ₂ O ₃	1.24	1.92	17.07
MgO	1.41	3.96	1.39
Cl ⁻	17.74	9.37	NA

Source: [4] and [26]. Note: NA, not available.

diameter of 9 cm), control system and collection system. The thermal chamber was made of steel (AISI 316) and utilized an electrical heater. It had both a temperature control system and a rotary kiln speed controller. The temperature control system had two thermocouples and a proportional integral derivative (PID) controller for temperature adjustment. The rotary kiln speed controller adjusted rotary kiln speed. The collection system was made of steel (AISI 316) and collected the thermally treated slag that fell from the rotary kiln.

2.3. Experimental procedure

The experimental procedure had some steps. The MSWI fly ash was washed with distilled water for 3 h in agitated vessels under room temperature (25 °C). The volume of agitated vessels was 2 L and the agitation rate was 30 rpm. The mass ratio of ash/water was 1:10. After washing, the solid/water mixture was separated via a glass fiber filter. The remaining solids were dried overnight in an oven at 105 °C. This part of the fly ash sample was classified as washed fly ash, and the remaining portion of MSWI fly ash was called raw fly ash. Second, MSWI bottom ash was added to both washed and raw fly ash in various ratios and mixed well. Three ratios of MSWI fly ash (*F*) to bottom ash (*B*), *B/F* = 0, 0.1 and 1, were used. Finally, the ash and mixed ash were fed into the thermal chamber under operational conditions. The operational temperature was 700 °C and the parameters included the form of pretreatment, the proportion of bottom ash (bottom ash/fly ash, *B/F* = 0, 0.1 and 1) and the three different retention time. The three different rotation rates included 0.89 rpm, 1.39 rpm and 2.0 rpm in this study. The detail operational conditions of this experiment were similar to those used by Chou et al. [25].

2.4. Sequential extraction procedure

The sequential extraction procedure was a modified version of that developed by Tessier et al. [11]. The equipment used for the sequential extraction procedure was 50 mL polyethylene (PE) vessel and into which the sample (2.5 g) was put. The sample and extraction solution were mixed by using a rotation machine and the heating equipment was water bath. All of the extraction conditions were summarized in Table 2. After extracting each fraction, the extraction solution was centrifuged (3000 rpm, 15 min) and filtered. The metal concentrations of each fraction were analyzed by flame atomic absorption spectroscopy (FAAS).

3. Result and discussion

3.1. Metal fraction in raw ash and washed ash

3.1.1. Distribution of Pb pattern

Fig. 1(a) shows the distribution of the Pb pattern in raw ash and washed ash. The main fraction in raw fly ash was carbonate (about 56%). The secondary fraction of raw fly ash was Fe–Mn oxides (about 21%). According to the experimental results, the lead fractions of

Table 2
Sequential extraction procedures

Fraction	Extraction agent	Extraction condition	
		Mixing time (h)	Temperature (°C)
Exchangeable	20 mL 1 M MgCl ₂	1	25
Carbonate	20 mL 1 M NaOAc	5	25
Fe–Mn oxides	50 mL 0.04 M NH ₂ OH·HCl in 25% HOAc	5	96 ± 3
Organic matter	7.5 mL HNO ₃ + 20 mL 30% H ₂ O ₂	5	85 ± 2
	12.5 mL 3.2 M NH ₄ OAc in 20% HNO ₃	0.5	25
Residual	25 mL aqua regia (HNO ₃ :HCl = 1:3)	8	80 ± 5

raw fly ash centered on carbonate and Fe–Mn oxides. The reason for this phenomenon can be explained by the fact that the Pb in the solid waste was vaporized during incineration and Pb vapor was condensed on the MSWI fly ash in the air pollutant control device. Because this condensation reaction happened very fast, Pb vapor could not entirely enter the lattice of MSWI fly ash entirely. Therefore, the chief fraction of Pb exhibited carbonate and Fe–Mn oxides. The primary fraction of washed raw fly ash also exhibited carbonate (30%) and Fe–Mn oxides (40%). Comparison of fractions in raw and washed raw fly ash indicated that the percentages of carbonate and Fe–Mn oxides differed. The percentage of carbonate fraction of Pb in washed raw fly ash decreased and the percentage of Fe–Mn oxides of Pb in washed raw fly ash was increased. These phenomena resulted from the washing process washing out some

parts of exchangeable fraction and carbonate fraction, while Fe–Mn oxides fraction was not washed out. Therefore, the Fe–Mn oxides fraction of Pb in washed raw fly ash was increased.

3.1.2. Distribution of Cu pattern

Fig. 1(b) displays the distribution of the Cu pattern in raw ash and washed ash. The predominant fraction in raw fly ash was carbonate (40%). The secondary fractions were Fe–Mn oxides (27%) and organic matters (28%). The pattern distribution of Cu was similar to that of Pb. Comparison of Pb and Cu in raw fly ash, it could be found that the percentage of organic matter fraction in Cu was higher than that in Pb. The experimental results for washed raw fly ash indicated that the fraction distribution of Cu was almost the same as that for raw fly ash. However, the percentage of Fe–Mn oxides fraction in washed raw fly ash was higher than that in raw fly ash. The difference may be due to the washing process influencing and reducing the percentage of exchangeable fraction and carbonate fraction without affecting the percentage of Fe–Mn oxides fraction. On the other hand, the percentage of Fe–Mn oxides fraction in Cu increased, and the distributions of organic matter and residual fraction of Cu in washed raw fly ash were similar to in raw fly ash.

3.1.3. Distribution of Cd pattern

Fig. 1(c) plots the distribution of the Cd pattern in raw ash and washed ash. The dominant fraction in raw fly ash was exchangeable (59%) and the secondary fraction was carbonate (35%). Because cadmium is more volatile than copper and lead, the dominant fractions of Cd were exchangeable fraction and carbonate fraction. In the washed raw fly ash part, the main fractions were also exchangeable fraction (30%) and carbonate fraction (51%). Comparison of the fraction of raw fly ash and washed raw fly ash indicated that the percentage of carbonate in washed raw fly ash was higher. As the washing process leached some parts of Cd in the exchangeable fraction, the percentage of exchangeable fraction decreased. However, the washing process did not influence the percentage of carbonate fraction significantly. Therefore, the increasing of exchangeable fraction in Cd only occurred in raw fly ash.

3.2. Effect of thermal treatment

3.2.1. Distribution of Pb pattern

Fig. 2(a) plots the distribution of the Pb pattern in raw ash and thermally treated raw fly ash slag. Comparison of the Pb fractions in raw fly ash and thermally treated slag indicated that these samples had a different distribution of Pb fraction. The main fraction in raw fly ash was carbonate, and the main fraction in thermally treated raw fly ash was Fe–Mn oxides. The percentages of Fe–Mn oxides and organic matter in thermally treated raw fly ash slag were 79% and 14%, respectively. This phenomenon resulted from some Pb species could be changed from carbonate to Fe–Mn oxides after thermal treatment. The thermal treatment also caused the pattern of Pb fraction to stabilize. However, the distribution of Pb

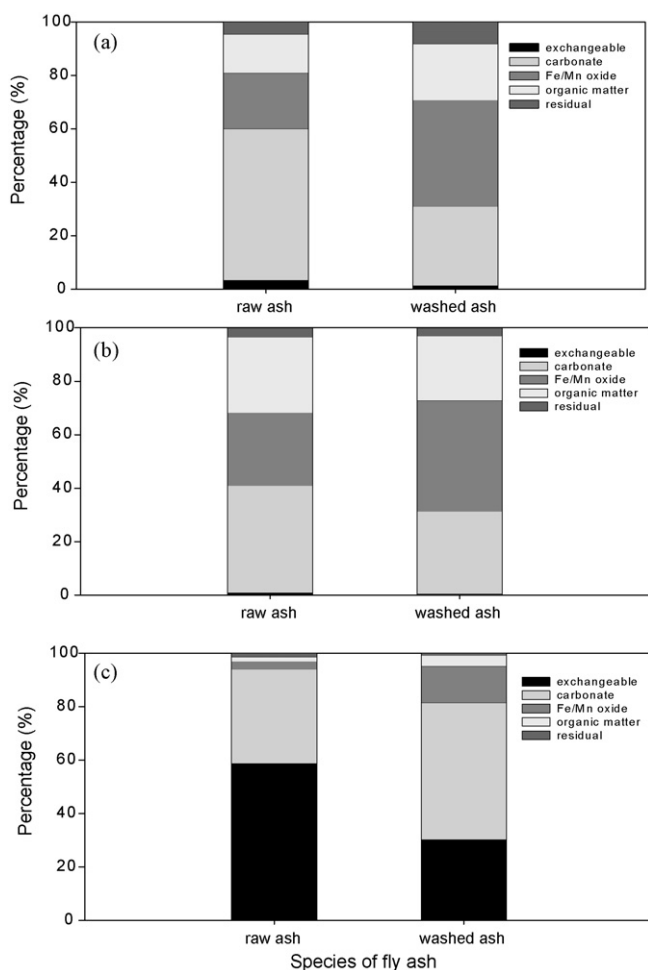


Fig. 1. The metals' distribution pattern in raw ash and washed ash: (a) Pb, (b) Cu and (c) Cd.

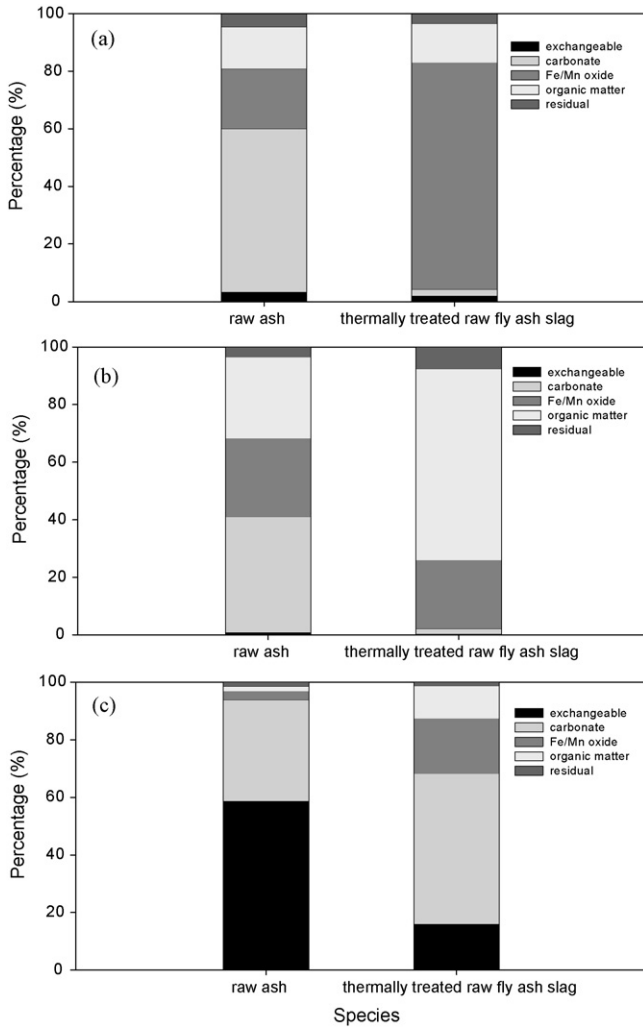


Fig. 2. The metals' distribution pattern in raw ash and thermally treated raw fly ash slag: (a) Pb, (b) Cu and (c) Cd.

in MSWI fly ash was converted to a stable fraction with thermal treatment.

3.2.2. Distribution of Cu pattern

Fig. 2(b) illustrates the distribution of the Cu pattern in raw fly ash and thermally treated raw fly ash slag. Raw fly ash and thermally treated raw fly ash slag differed in that the main fraction in raw fly ash was carbonate fraction, while that of thermally treated raw fly ash slag was organic matter (66%). According to experimental results, the distribution of Cu changed after thermal treatment. The reason for the conversion of the Cu pattern may be due to the oxidation reaction occurred with thermal treatment. The oxidation reaction also occurred in the other two metals. In conclusion, MSWI fly ash treated thermally reduce the leachability of Pb and Cu.

3.2.3. Distribution of Cd pattern

Fig. 2(c) shows the distribution of the Cd pattern in raw ash and thermally treated raw fly ash slag. Comparison of the main fractions in raw fly ash and thermally treated raw fly ash slag showed that the predominant fraction in raw fly ash were exchangeable fraction and carbonate fraction, while the fraction in thermally treated raw fly ash slag was carbonate fraction (55%). In addition to increase in carbonate, the percentages of the Fe–Mn oxides and organic matter in thermally treated raw fly ash slag also increased. Thermal

treatment did not only influence stable metal but also on unstable metal. As Cd is an unstable metal, the distribution of the Cd pattern was also stabilize.

3.3. Effect of pretreatment

3.3.1. Distribution of Pb pattern

Fig. 3(a) shows the distribution of the Pb pattern in thermally treated raw fly ash slag and thermally treated washed fly ash slag. Comparison of these distributions demonstrated that the percentage of exchangeable fraction and carbonate fraction in thermally treated raw fly ash slag and thermally treated washed fly ash slag were almost the same. The percentage of Fe–Mn oxides in thermally treated raw fly ash slag and washed fly ash slag were 79% and 57%, respectively. The percentage of organic matter fraction in thermally treated raw fly ash slag was 14% and in washed fly ash slag was 36%. According to the distribution of these slags, the washing process reduced the percentage of Fe–Mn oxides and increased the percentage of organic matter in thermally treated washed fly ash slag. However, pretreatment of thermally treated slag affected in Fe–Mn oxides and organic matter fractions.

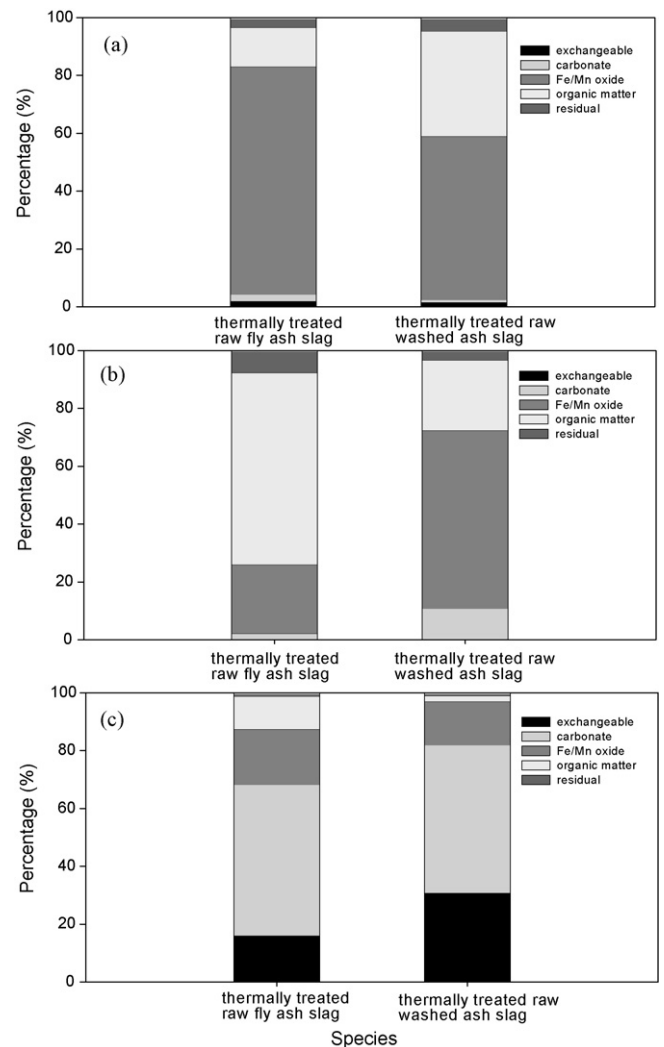


Fig. 3. The metals' distribution pattern in thermally treated raw fly ash slag and thermally treated washed fly ash slag: (a) Pb, (b) Cu and (c) Cd.

3.3.2. Distribution of Cu pattern

Fig. 3(b) depicts the distribution of the Cu pattern in thermally treated raw fly ash slag and thermally treated washed fly ash slag. The distribution of major fractions was organic matter (66%) and Fe–Mn oxides (61%) for thermally treated raw fly ash slag and washed fly ash slag, respectively. The percentages of the other three fractions in these two slags were not so high. The effect of pretreatment in thermally treated washed fly ash slag was the change in fraction. The washing process changed the distribution of Cu and converted organic matter into Fe–Mn oxides. However, the effect of pretreatment in distribution of the Cu pattern was significant.

3.3.3. Distribution of Cd pattern

Fig. 3(c) shows the distribution of the Cd pattern in thermally treated raw fly ash slag and thermally treated washed fly ash slag. The main fraction in thermally treated raw fly ash slag was carbonate fraction (52%). The secondary fractions of thermally treated raw fly ash slag were exchangeable fraction (16%) and Fe–Mn oxides fraction (19%). Carbonate fraction (51%) remained the main fraction in thermally treated washed fly ash slag. Comparison of these two slags indicated that the percentage of Fe–Mn oxides and organic matter fraction in thermally treated washed fly ash slag had decreased slightly. However, the fraction distribution in thermally treated raw fly ash slag and washed fly ash slag did not change significantly.

3.4. Effect of retention time

3.4.1. Distribution of Pb pattern

Fig. 4(a) displays the distribution of the Pb pattern in thermally treated raw fly ash slag with different retention time. At the shortest retention time (2.0 rpm), the main fraction was Fe–Mn oxides (75%). The secondary fraction was organic matter (11%). At a moderate retention time (1.39 rpm), Fe–Mn oxides (78%) remained the main fraction with organic matter (11%) still the secondary fraction. At the longest retention time (0.89 rpm), Fe–Mn oxides were 63% and organic matter (29%), the secondary fraction was 29%. As retention time increased, the Pb patterns became increasingly stable. Although the percentage of Fe–Mn oxides decreased at the longest retention time, the percentage of organic matter increased. In conclusion, retention time significantly influenced the distribution of Pb patterns.

3.4.2. Distribution of Cu pattern

Fig. 4(b) depicts the distribution of the Cu pattern in thermally treated raw fly ash slag with different retention time. At the longest retention time (0.89 rpm), the main fraction was Fe–Mn oxides (72%) and the secondary fraction was organic matter (23%). At a moderate (1.39 rpm) and shortest retention time (2.0 rpm), the main and secondary fractions were similar to those at longest retention time. As retention time increased, the percentage of organic matter increased. The reason for this phenomenon may be that retention time was related to the Cu pattern. As retention time increased, the Cu pattern became increasingly stable. In conclusion, the distribution of Pb and Cu patterns with different retention time were roughly the same because Pb and Cu were the stable metals.

3.4.3. Distribution of Cd pattern

Fig. 4(c) shows the distribution of the Cd pattern in thermally treated raw fly ash slag with different retention time. The two major fractions of distribution patterns in Cd were carbonate and Fe–Mn under different retention time. The main fraction was carbonate (46%) and the secondary fraction was Fe–Mn oxides (24%) when retention time was the shortest. The percentage of carbonate and Fe–Mn oxides with a moderate retention time were 59% and 16%,

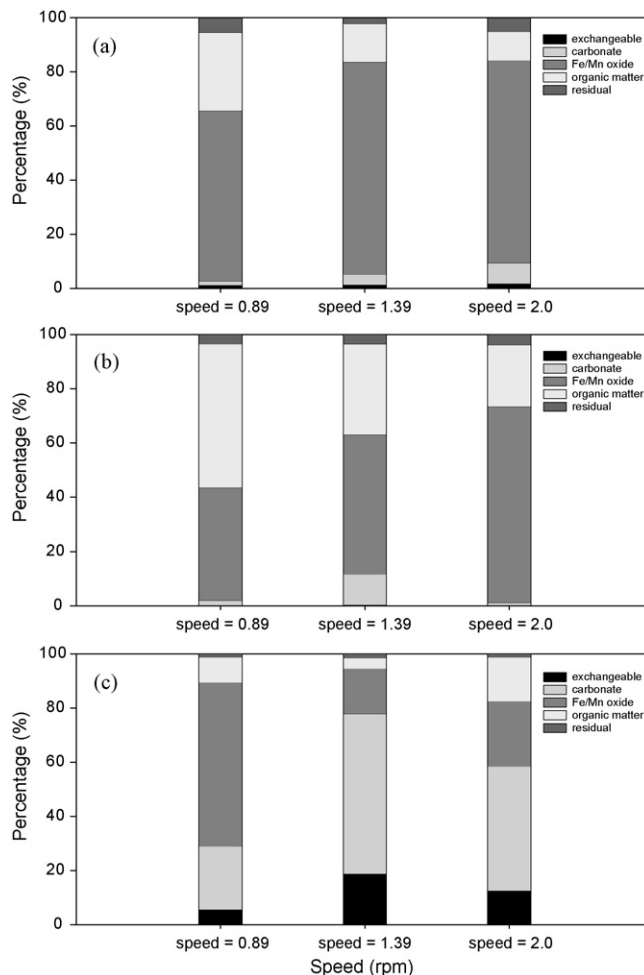


Fig. 4. The metals' distribution pattern in thermally treated raw fly ash slag with different retention time: (a) Pb, (b) Cu and (c) Cd.

respectively. When the retention time was longest, the percentage of carbonate and Fe–Mn oxides were 23% and 61%, respectively. Comparison of distribution with different retention time indicated that as retention time increased, the pattern distribution of Cd became increasingly stable. However, the retention time had a significant influence on the conversion of the Cd fraction.

3.5. Effect of additive ratio

3.5.1. Distribution of Pb pattern

Fig. 5(a) shows the distribution of the Pb pattern in thermally treated slag with different amounts of bottom ash. The main fraction in thermally treated slag was Fe–Mn oxides (78%) when $B/F=0$. At $B/F=1$, the main fractions in thermally treated were Fe–Mn oxides (42%) and organic matter (43%). At $B/F=0.1$, the main fraction in thermally treated slag was Fe–Mn oxides (78%). According to experimental results, the distribution of the Pb pattern in thermally treated slag was very similar at $B/F=0$ and 0.1. The distribution of the Pb pattern in thermally treated slag did not differ significantly when a small amount of bottom ash was added. However, at $B/F=1$, the distribution of Pb in thermally treated slag had obvious variation. This variation resulted from the fact that metal in bottom ash was more stable than that in fly ash. In conclusion, adding sufficient amounts of bottom ash could stabilize the Pb patterns.

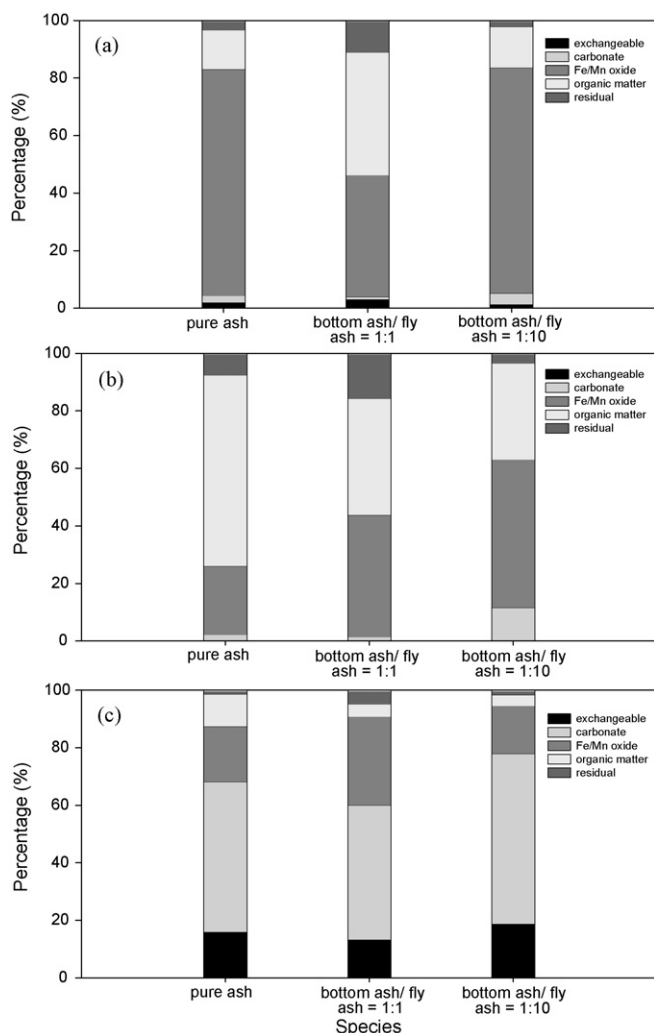


Fig. 5. The metals' distribution pattern in thermally treated slag with different amounts of bottom ash: (a) Pb, (b) Cu and (c) Cd.

3.5.2. Distribution of Cu pattern

Fig. 5(b) displays the distribution of the Cu pattern in thermally treated raw fly ash slag with different amounts of bottom ash. The main fraction in thermally treated slag was organic matter (66%) when $B/F=0$. At $B/F=0.1$, the main fractions in thermally treated slag were Fe–Mn oxides (42%) and organic matter (40%). At $B/F=0.1$, the main fraction was Fe–Mn oxides (51%). Experimental results showed that the percentage of Fe–Mn oxides increased and the percentage of organic matter decreased when bottom ash was added. However, adding bottom ash could increase the percentage of Fe–Mn oxides of Cu with a potential adverse environmental impact.

3.5.3. Distribution of Cd pattern

Fig. 5(c) illustrates the distribution of the Cd pattern in thermally treated raw fly ash slag with different amounts of bottom ash. The predominant fraction in thermally treated slag was carbonate (52%) when $B/F=0$. At $B/F=1$, it was still carbonate (46%) but the percentage of carbonate had decreased and that of Fe–Mn oxides had increased. At $B/F=0.1$, the main fraction was carbonate (59%). Although the main fraction in these slags were roughly the same, the percentage of Fe–Mn oxides increased when $B/F=1$. In conclusion, adding MSWI bottom ash could increase the stability of the Cd pattern in thermally treated slag.

4. Conclusion

Although MSWI fly ash is a hazardous material, its reuse potential had been increased by thermal treatment. However, methods to change it into a non-hazardous material and reduce its leaching concentration of MSWI fly ash are very important topic. Thermal treatment of MSWI fly ash could reduce its metal leaching concentrations of MSWI fly ash effectively. This study evaluated the Pb, Cu and Cd patterns in MSWI fly ash during thermal treatment. Different parameters were studied and the following are the main conclusions.

1. In raw fly ash, the main fraction in Pb and Cu was carbonate and that in Cd was exchangeable. This experimental finding is because that Pb and Cu are stable metals while Cd is less stable and highly mobile. In washed fly ash, the main fraction in Pb and Cu was Fe–Mn oxides and the fraction in Cd was carbonate. The distribution of the Pb, Cu and Cd patterns were different in raw fly ash and washed ash. The reason for these differences could be explained by the fact that washing removed some soluble metals. Therefore, the distribution of Pb, Cu and Cd in raw fly ash and washed fly ash differed.
2. The distributions of Pb, Cu and Cd with and without thermal treatment differed. In thermally treated slag, the main fraction in Pb was Fe–Mn oxides. The main fraction in Cu was organic matter, and that in Cd was carbonate. The high-temperature environment in thermal treatment stabilized the fractions of Pb, Cu and Cd. Thus, the thermal treatment reduced the leaching concentration in thermally treated slag.
3. Washing influenced the distribution of Pb and Cu in thermally treated slag, but did not influence the distribution of Cd. Washing pretreatment increased the percentage of organic matter in Pb and reduced percentage of organic matter in Cu. Therefore, the washing pretreatment only stabilized Pb.
4. The distribution patterns of Pb, Cu and Cd stabilized when retention time increased. Comparison of different time showed that the percentage of Fe–Mn oxides in Pb and Cu decreased and the percentage of organic matter in Pb and Cu increased when retention time increased. Even though Cd has a high mobility, the percentage of Fe–Mn oxides in Cd also increased as retention time increased. Therefore, the retention time could increase the percentage of stable fraction in Pb, Cu and Cd, meaning that the potential adverse impact on the environment was decreased.
5. Adding different amounts of bottom ash also affected the distributions of Pb, Cu and Cd. Comparison of raw fly ash and thermally co-treated slag demonstrated that the distribution patterns of Pb, Cu and Cd differed, especially when $B/F=1$. When $B/F=1$, the percentage of organic matter in Pb as well as the percentage of Fe–Mn oxides in Cu and Cd increased. Therefore, adding different amounts of bottom ash effectively influenced on Pb and Cd patterns; the effect on distribution of Cu was not significant.

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