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Thermal treatment of the fly ash from municipal solid waste incinerator with rotary kiln

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Abstract

Reuse of the fly ash from the municipal solid waste incinerator (MSWI) is a policy of Taiwan EPA. However, the fly ash is often classified as a hazardous waste and cannot be reused directly because the concentrations of heavy metals exceed the TCLP regulations. The main objective of this study is to investigate the continuous sintering behavior of fly ash with a rotary kiln and seek a solution to reduce the concentrations of heavy metal to an acceptable value. The partitions of the heavy metals in the process are also considered.

The results of TCLP showed that among the metals of Cr, Cd, Cu and Pb, only the concentrations of Pb in raw fly ash exceeded the regulation. At sintering temperatures of 700, 800 and 900 °C, the concentration of Pb decreased in sintering products, however, the concentration of Pb still exceeded the limitation at 700 and 800 °C. Additionally, the water-washing was used to pre-treat the fly ash before sintering process. The washing treatment effectively reduced the leaching concentrations of Pb to agree the regulations. Therefore, water-washing followed by a sintering treatment is an available process for detoxifying the fly ash of MSWI.

Keywords: Fly ash; MSWI; Sintering; Heavy metal; Rotary kiln

1. Introduction

The municipal and industrial solid wastes have increasing with years in Taiwan. Incineration is applied as a major treatment before the final landfill to prolong the life of the landfill. During incineration, the air pollutants such as fly ash, organic compounds and acid gases, needs to be removed from exhaust gas to agree the emission standards. Heavy metals with higher saturated vapor pressure, such as Hg, Cd and Pb, are easily volatilized into the flue gas, which accounts for their high content in fly ash. However, the collected fly ash is classified as the hazardous waste because the TCLP leaching concentrations of heavy metals do not agree the regulations. Therefore, a treatment process must be used to reduce the TCLP concentration of the fly ash to avoid the second pollution when fly ash is reused.

Generally, the main compositions of fly ash are Si, Al, Ca, K, Na, Cl, etc. [1]. MSWI fly ash has smaller particle size and density than bottom ash, and higher ability of absorbing water [2]. As aforementioned, toxicity of fly ash generates from the heavy metal and its compounds. During incineration, the metal species in the waste can transform between the different types but they cannot be destroyed. When the flue gas cools down, metal species will subsequently condense to from metallic particles. These toxic heavy metals may escape from the process in molten slag, dry collected ash, wet scrubber ash, or as an airborne aerosol exiting the stack [3–5].

The major approaches of handling MSWI fly ash include solidification/stabilization, melting and sintering. Solidification/stabilization adds additives to remove the water in the waste, and then the hazardous materials encapsulated or bonded inside of the agglomerates. The additives may react with specific compositions in the waste to form the more harmful matters, then released into the environment if the solidity breaks. When the melting process is used, the organic materials of fly ash are decomposed, gasified and combusted under high operating temperature. In addition, the inorganic compounds become slag, and the hazardous materials are encapsulated by the Si–O structure of the slag [3]. The emissions of heavy metals during thermal treatment are affected by the components of substance, reaction atmosphere and reaction temperature. With rich oxygen, metal

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oxides are the dominant species; while volatile metal chlorides are easily formed, instead of metal oxides, when the system contains more chlorine. When sulfur is present, metal sulfates are the major species at low temperature and metal oxides or metal chlorides are dominant at high temperature [5–9]. Furthermore, the emissions of heavy metals are increased with the operating temperature [10–12]. Díaz-Somoano et al. [13] addressed the emission of heavy metals was also affected by reaction atmosphere. In oxidizing condition, metal oxides were formed easily. Some complex compounds, such as metal sulfides, were formed in reducing condition, depending on the compositions of materials [14].

The operation temperature of sintering treatment is lower than the melting point of wastes, the sintering temperature is usually 700–1100 °C. It is classified in two kinds of process including liquid phase sintering and solid phase sintering. There have different mobile modes of particles and atoms [15–17]. The sintering treatment has the advantages of reducing weight, volume and encapsulating the hazardous materials which is the same as a melting process, and even lower operation cost and technology requirements than melting. Moreover, the physical/chemical characteristics of sintering products are better than the products of solidify [15,16]. The qualities of the sintering products, such as mechanical strength, density, shape, etc., are influenced by the operating parameters of temperature, rising rate of temperature and retention time. The physical and chemical characteristics of the feeding materials, including components of ash, particle size and additives, etc., also affect the qualities of the sintering products [18,19]. There are some minor compounds like alkali metal, alkaline-earth metal, chloride and sulfate existed in fly ash, the partitions of different compounds influence the temperature of the sintering operation and the quality of products [20–22].

Researches of the sintering treatment now are focused on reuses of the fly ash, the sludge from sewers, water treatment plants or reservoirs in Taiwan. The general applications of sintering products are used as the raw materials for the lightweight aggregate, cement and permeable or compressive pavements [23–28]. These studies are concerned with the engineering characteristics of the sintering products. Some other researchers focus on sintering operations of the fly ash, discussing about evaporation of heavy metals, changes of metal compounds or leaching of sintering products [7,29,30]. However, the experiments of these studies are carried out in batch operations, using tube furnaces to heat the fly ash. A batch system needs a storage tank and conveying facilities when it is applied to treat the MSWI. These methods will increase the complexity and cost.

A continuous sintering process using a rotary kiln with continuous feeder system is designed to sinter the fly ash for encapsulating the heavy metals. The rotary kiln is widely employed because it has the advantages of good mixing and continuous operation. It can be easily fitted to the exit of the air pollution control devices to operate continuously. The heavy metals will exist unavoidably in the exhaust gas of the rotary kiln under high temperature, so the partitions of heavy metals between the sintered fly ash and exhaust gas needs to be understood. The operating parameters of rotary kiln include retention time, temperature, rotary speed and tilt angle of rotary kiln, etc. In this study, the fly ash of MSWI was used to discuss and the retention time and temperature were selected because they were main factors to affect the properties of sintering products. The other parameters were fixed to compare the results of raw materials and their sintered products. The main purpose of this study was finding a practical method to reduce the leaching of the heavy metals to agree the hazardous waste regulations. The pretreatment of the fly ash by water-washing was also used to evaluate the possibilities of lowering the TCLP concentration or sintering temperature.

Table 1
Particle size distribution (wt.%) of MSWI fly ash

Particle size range	Individual ratio (%)	Accumulative ratio (%)
>425 μm	0.06	0.06
250–425 μm	1.35	1.41
180–250 μm	3.07	4.48
125–180 μm	3.96	8.44
90–125 μm	4.95	13.39
75–90 μm	5.17	18.56
38–75 μm	57.16	75.72
<38 μm	24.28	100.00

2. Experimental

2.1. Materials

In this study, the fly ash was collected from a mass-burning incinerator in Taiwan. The particle size distribution of the fly ash was listed in Table 1, only few particles were above 425 μm (0.06%) and more over 50% of the particles were in the range of 38–75 μm. Parts of the fly ash were washed by water to be water-washing ash then the fly ash and washed fly ash were sintered to serve as the sintered fly ash and sintered water-washing ash, respectively. These four types of fly ashes were identified in this study.

2.2. Apparatus

Fig. 1 depicts the sintering system. The rotary kiln incinerator consisted of a continuous feeder system, a main sintering chamber (210 cm in length, 9 cm in i.d.) and a collection system

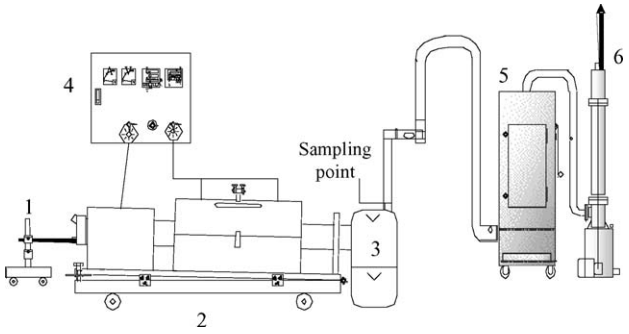

Fig. 1. The rotary kiln incinerator: (1) feeding system, (2) rotary kiln incinerator, (3) collection system, (4) control system, (5) air pollution control system, and (6) stack.

Table 2
Operating conditions of the experiment

Run no.	Sintering temperature (°C)	Sintering time (h)	Feedstock
1	700	1	MSWI fly ash
2	700	1.5	
3	700	2	
4	800	1	
5	800	1.5	
6	800	2	
7	900	1	
8	900	1.5	
9	900	2	
11	700	1	Water-washing fly ash
12	700	1.5	
13	700	2	
14	800	1	
15	800	1.5	
16	800	2	
17	900	1	
18	900	1.5	
19	900	2	

of sintered ash. The chamber was made of stainless steel (AISI 316). One thermocouple was used to monitor the temperature in the chamber. The exhaust gas was cleaned by air pollution control device then released into the atmosphere. The temperature and rotary speed of the chamber were regulated by the control system. The electrical heater linked to a proportional integral derivative (PID) controller was used to heat the rotary kiln.

2.3. Experimental procedure

The fly ash collected from a MSWI was mixed well at first, and then parts of the fly ashes were washed with distilled water. The washing time was 3 h and the solid/water ratio was 1/20. After water-washing, the solid and liquid were separated and the solid were dried at 105 °C for 24 h. Then the fly ash and water-washing ash were sintered under different temperatures and retention time separately. The operation temperatures were 700, 800 and 900 °C, and the retention time were 1, 1.5 and 2 h. Table 2 lists the experimental conditions of this study. The rotary kiln was rotating in low speed to avoid the main chamber crack when it was heated firstly. After the temperature reaching the steady state, rotary speed of the kiln was raised to the normal condition, fly ashes were fed in the rate of 0.7 g/s and the sintering product was collected to analyze. The TCLP concentration of the fly ash and sintering products were analyzed by flame atomic absorption spectroscopy (F.A.A.S.). The surface structure and the elements of the fly ash and products were determined by SEM and XRD. Particulates and gas pollutant were sampled from the exhaust gas to analyze the metal partitions. The sampling procedure follows the standard method "NIEA A302.72C" of Taiwan EPA. The solid samples, including the sintering products and the particles on the filter, were extracted by microwave digestion. All the samples were analyzed by F.A.A.S.

Table 3
Basic compositions of fly ash

Composition	Ratio (%)
SiO_2	7.62
Al_2O_3	3.38
Fe_2O_3	1.24
CaO	35.81
MgO	1.41
SO_4	1.35
Cl^-	17.74
Cr (mg/kg)	103.59
Cd (mg/kg)	57.84
Cu (mg/kg)	359.44
Pb (mg/kg)	304.33

3. Results and discussion

3.1. Characteristics of the fly ash

The fly ash in this experiment was collected a MSWI in Taiwan. The bulk density is 300.97 kg/m^3, and the loss of ignition is 0.27%. The chemical compositions of the fly ash and water-washing ash, including CaO, Cl^-, SiO_2, and Al_2O_3, were listed in Tables 3 and 4, respectively. The large quantity of CaO arises from the air pollution control device, adding the solution of CaO in the spray dryer to react with the acid gas. The high concentration of the chlorine in the solid waste increased the ratio of Cl^- in the fly ash. The water-washing treatment reduced the chlorine content from 17.7 to 9.4% after the fly ash was washed.

3.2. TCLP leaching concentration of the fly ash and sintering products

Fig. 2 shows the TCLP leaching concentrations of Cr, Cd, Cu and Pb in the fly ash and sintered fly ash under different operating temperatures and durations. The leaching concentrations of the target metals in the fly ash agree the requirements of the regulation, except of Pb. Therefore, the fly ash could be reused unless the concentration of Pb was decreased below limitations. The TCLP leaching concentrations of Cr of the sintered fly ash was higher than those of the feedstock ash in all sintering conditions, as shown in Fig. 2(a). This result was different from the other metals. The reason of high TCLP concentration of the sin-

Table 4
Basic compositions of water-washing ash

Composition	Ratio (%)
SiO_2	15.15
Al_2O_3	7.29
Fe_2O_3	1.92
CaO	41.54
MgO	3.96
Cl^-	9.37
Cr (mg/kg)	128.76
Cd (mg/kg)	92.4
Cu (mg/kg)	608.41
Pb (mg/kg)	1034.7

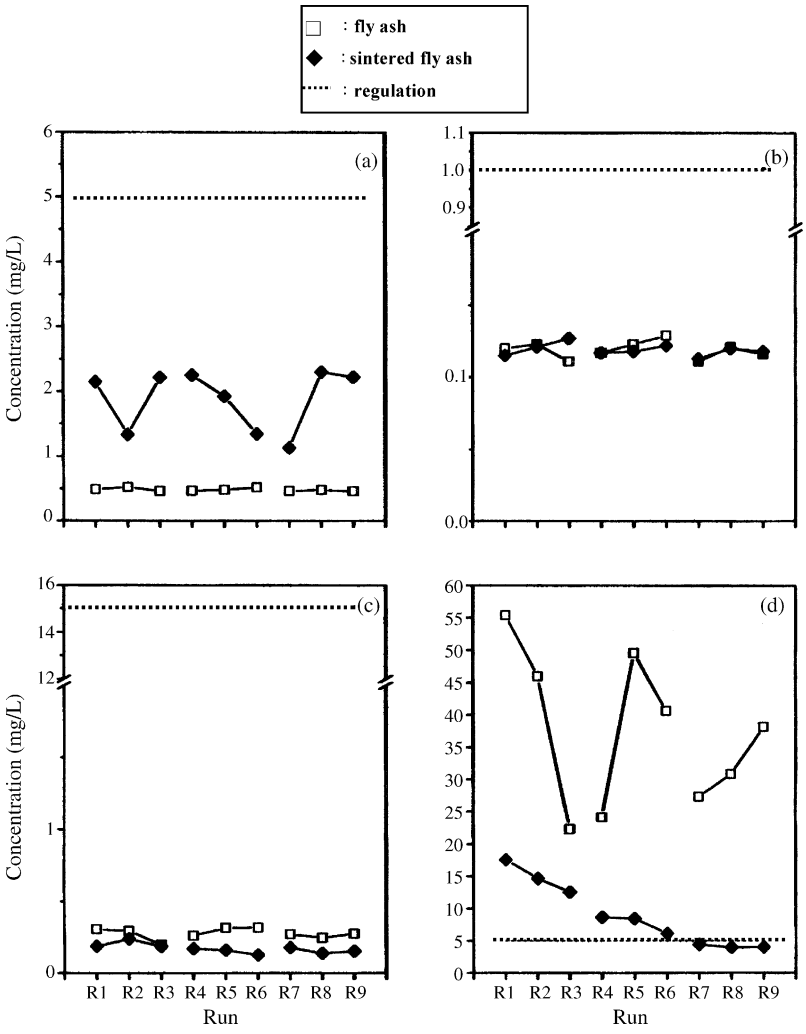

Fig. 2. TCLP concentrations of the fly ash and the sintered fly ash: (a) Cr, (b) Cd, (c) Cu, and (d) Pb.

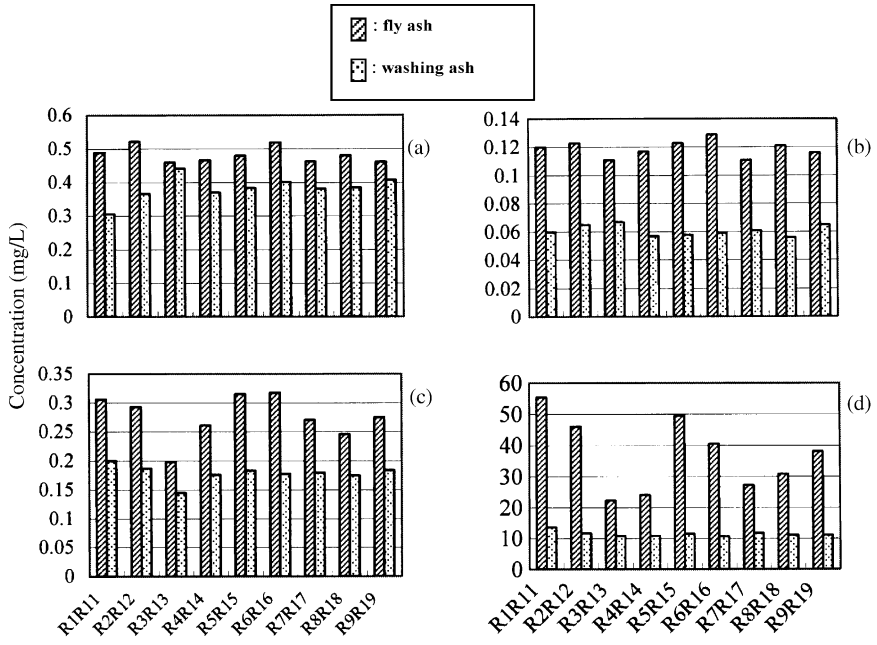

Fig. 3. TCLP results of the fly ash and water-washing ash: (a) Cr, (b) Cd, (c) Cu, and (d) Pb.

tering products might be that the species of Cr was transformed to the compounds, which were easily to be extracted [31]. However, The TCLP leaching concentrations of the sintered fly ash were still under the regulation of Cr. The sintering treatment seemed having little effect in reducing the leaching concentration of Cd, as shown in Fig. 2(b). The TCLP concentrations of Cu decreased after the fly ash was sintered under different conditions, as shown in Fig. 2(c). The mean reduction was about 36%. The concentration of Pb in sintered fly ash was lower than the fly ash. The reductions for the sintering treatment ranged from 44 to 89%. The TCLP concentrations of the sintering products declined with the increasing operating temperature. When the temperature was 900 °C, the concentration was lower than the regulation which meant the sintering product is not a hazardous waste anymore.

There are four stages in sintering process, including adhesion, initial, intermediate and the final sintering. In the first and second stages, the behaviors of particle are contact-formation and neck growth. The sintering driving force will make particle size become small and inter-pore structure become smooth. Then the driving force makes grain growing and the outer opening between particles disappear in third stage. In final stage, the inner pore of the particle separates each other to form an individual pore without opening. The densification phenomenon of particle and particle density increasing are appeared in final stage. During sintering process, different species formed if there were some different elements existed [5]. With high chlorine content in the fly ash, metal chlorides were easily formed and evaporated due to their low boiling points. The structures of sintering products were loosen when metal chlorides were evaporated. Therefore, high content of chlorine in the fly ash enhanced the formation and evaporation of metal chlorides, as well as the leachability of heavy metals from the sintering products.

3.3. Sintering treatment of the water-washing fly ash under different conditions

As aforementioned, TCLP concentration of Pb of sintering product agreed the regulations under the sintering temperature of 900 °C. However, the products sintered at 700 and 800 °C

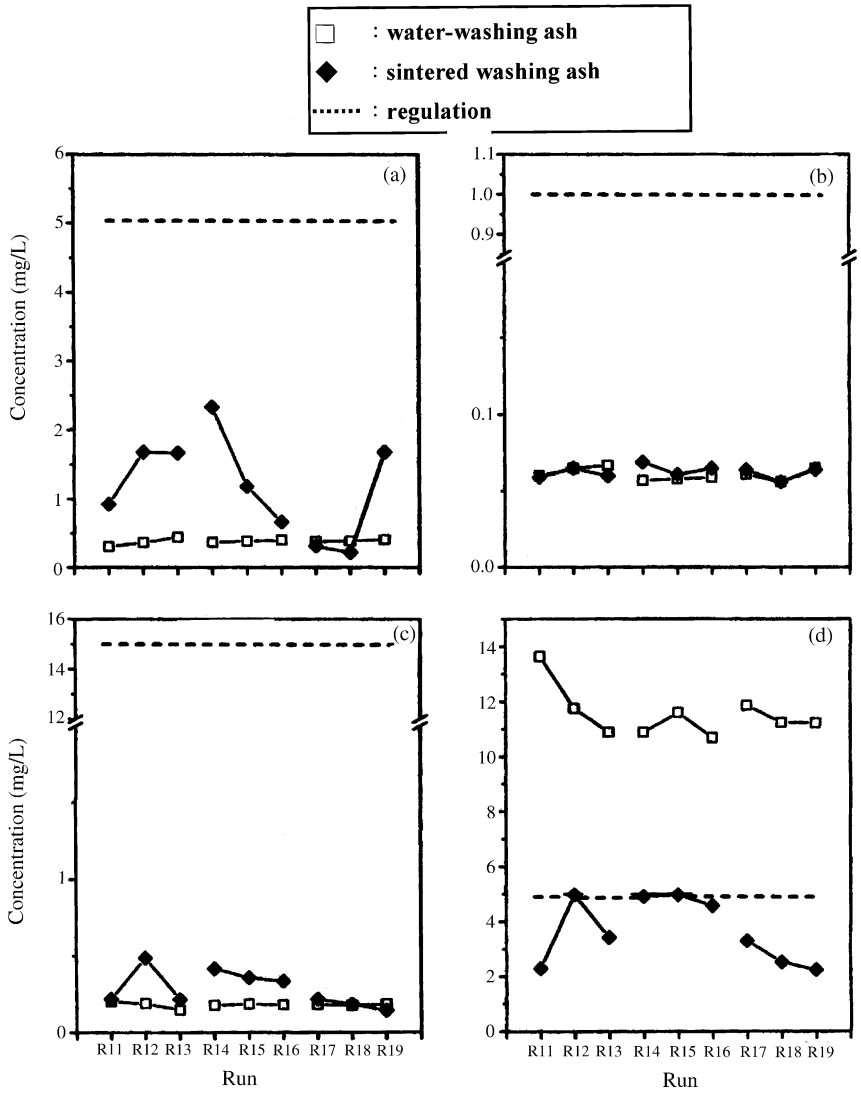

Fig. 4. TCLP concentrations of the water-washing ash and sintered water-washing ash: (a) Cr, (b) Cd, (c) Cu, and (d) Pb.

Table 5
Partitions of the various metals in the sintering process

Run no.	Cd			Cr			Cu			Pb		
	SP (%)	SS (%)	GP (%)	SP (%)	SS (%)	GP (%)	SP (%)	SS (%)	GP (%)	SP (%)	SS (%)	GP (%)
R1	99.51	0.41	0.08	99.85	0.15	0.00	99.87	0.12	0.01	99.66	0.31	0.03
R2	99.71	0.17	0.11	99.88	0.12	0.00	99.96	0.03	0.01	99.47	0.31	0.22
R3	99.47	0.46	0.07	99.98	0.02	0.00	99.75	0.25	0.00	97.03	2.95	0.02
R4	98.38	1.46	0.16	99.40	0.60	0.00	99.36	0.63	0.00	94.76	5.23	0.01
R5	97.42	2.42	0.16	99.26	0.74	0.00	98.85	1.15	0.00	92.35	7.63	0.03
R6	96.42	3.46	0.12	99.36	0.64	0.00	99.08	0.92	0.00	89.17	10.82	0.01
R7	97.14	2.71	0.15	98.41	1.04	0.55	98.61	1.39	0.01	95.82	4.16	0.02
R8	94.21	5.64	0.15	97.43	2.57	0.00	95.23	4.76	0.00	97.17	2.80	0.04
R9	92.70	6.84	0.46	91.85	8.14	0.00	95.36	4.61	0.03	84.25	15.68	0.06
R11	99.10	0.81	0.09	99.99	0.01	0.00	99.90	0.10	0.00	98.42	1.57	0.01
R12	99.61	0.29	0.10	99.99	0.01	0.00	99.83	0.16	0.00	97.72	2.27	0.01
R13	99.77	0.22	0.01	100.00	0.00	0.00	99.93	0.07	0.00	99.04	0.96	0.00
R14	99.39	0.57	0.04	99.98	0.02	0.00	99.60	0.40	0.00	93.79	6.21	0.01
R15	98.99	0.94	0.07	99.99	0.01	0.00	98.68	1.31	0.01	80.08	19.89	0.03
R16	99.64	0.33	0.03	100.00	0.00	0.00	99.18	0.82	0.00	91.18	8.81	0.01
R17	98.17	1.79	0.05	99.98	0.02	0.00	98.16	1.83	0.01	77.05	22.94	0.01
R18	98.68	1.27	0.05	99.95	0.05	0.00	98.05	1.95	0.00	85.30	14.69	0.01
R19	97.46	2.48	0.06	99.98	0.02	0.00	98.02	1.97	0.01	81.84	18.16	0.01

SP = sintering product; SS = solid phase; GP = gas phase; R1–R9 = raw fly ash; R11–R19 = washing ash.

were still classified as hazardous wastes. Mangialardi [28] used water to wash the fly ash and concluded that water-washing greatly improved the chemical and mechanical characteristics of sintering products. Therefore, water-washing was selected as a pre-treatment to lower the sintering temperature.

Fig. 3 compares the TCLP concentration of Cr, Cd, Cu and Pb in the raw fly ash with those of water-washing ash. The TCLP concentration of Cr, Cd, Cu and Pb decreased after fly ash was washed. Wang et al. [31] used water-extraction process to investigate the heavy metal behavior in fly ash. They found water-extraction could leach out some major elements and heavy metals, like Cl, K Na, Cr, Cd, Pb and Cu, etc. Zn, Cr, Cd and Cu content in fly ash were not too large, so the decreased trend was not significant even washing process was used. Due to the higher concentration of Pb in fly ash than Cr, Cd and Cu, the decreased concentration of Pb was significant after washing

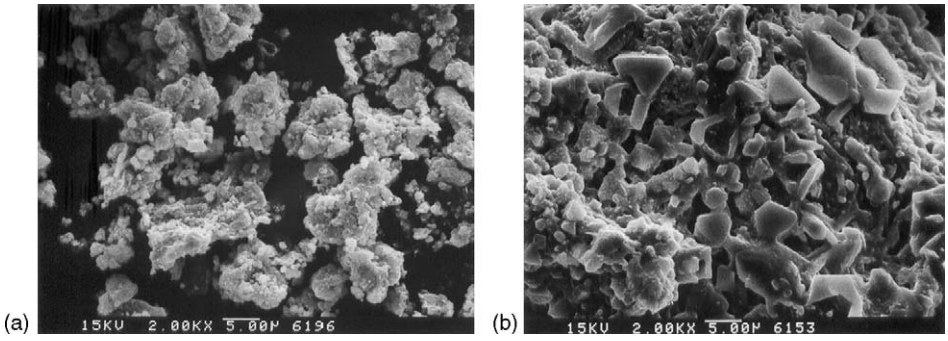

Fig. 5. SEM micrographs of the fly ash and sintering products (2000×): (a) feedstock of the fly ash and (b) sintered fly ash under temperature of 900 °C for 2 h.

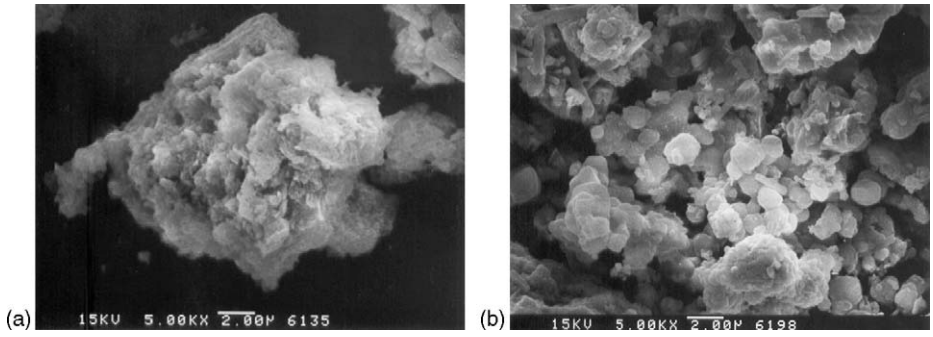

Fig. 6. SEM of water-washing ash and sintering products (5000×): (a) water-washing ash and (b) sintered water-washing ash under temperature of 900 °C for 2 h.

process. Before and after the washing treatment, Cr, Cd and Cu concentration of TCLP test were under the regulations. However, the Pb concentration of TCLP test was over regulation in both cases. Therefore, the water-washing ash is still the hazardous solid waste.

Fig. 4 shows the TCLP concentrations of Cr, Cd, Cu and Pb in water-washing fly ash and the sintered water-washing ash. The TCLP concentration of Cr in the sintered water-washing ash was higher than the concentration in the washing ash except of the temperature of 900 °C. The concentrations of Cd and Cu in the

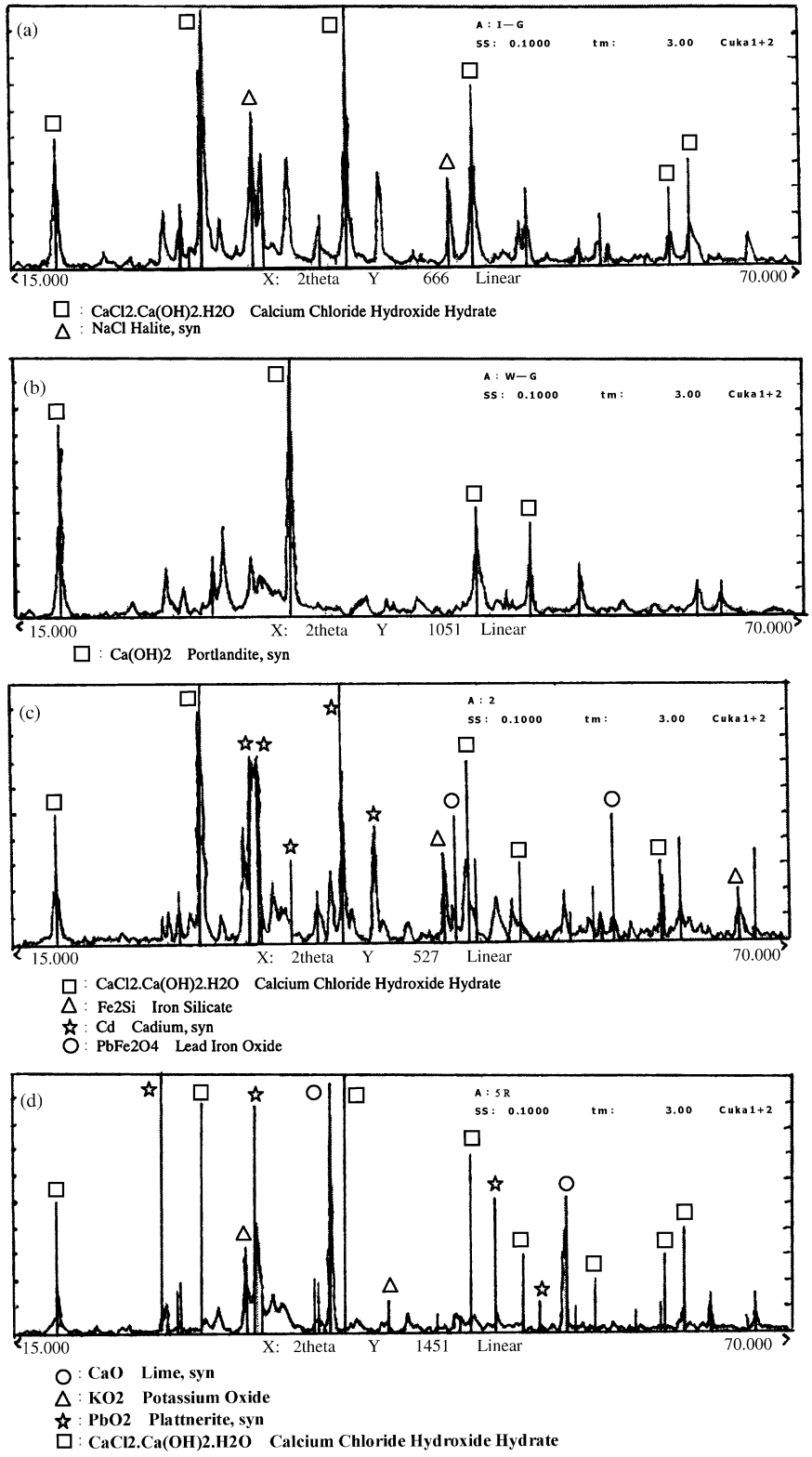

Fig. 7. XRD of fly ash, water-washing ash and sintering products (a) fly ash, (b) water-washing ash, (c) sintered fly ash, and (d) sintered water-washing ash.

sintered products slightly differed from those of the washing ash. Although the concentration of Cu of the product was higher than the concentration of water-washing ash, the absolute value and the difference of concentration were too low to be significant. As for Pb, the results of sintering treatment agreed the regulation at each operating condition of various temperature and duration. If the fly ash was washed, the sintering temperature could decline from 900 to 700 °C, and the fly ash became non-hazardous material. As reported in previous paper [28], modification of MSW fly ash by water-washing treatment could improve the chemical and mechanical characteristics of sintered products. The declined temperature of sintering treatment is a benefit to energy saving and air pollution control since more metal compounds will be evaporated under higher temperature.

3.4. Metal partitions among the fly ash, sintering product and the exhaust gas

When the fly ash was sintered under high temperature, the heavy metals might be encapsulated, vaporized or condensed, so the metal partitions changed. The air pollution occurred when the metals entered into the exhaust gas and were not removed by the air pollution control device. Therefore, the metal partitions were important information for the sintering treatment of the fly ash. Table 5 lists the partitions of the various metals of the fly ash and water-washing ash in the sintering process. The metals existed in the sintering product. The solid phase was collected by the filter and the gas phase was collected by the absorption liquid in the exhaust gas. The experimental data showed the metals existed mainly in the sintering products, and the ratios were higher than 80% for various metals under different conditions. The gas phase partitions of the four metals are low enough and can be neglected. The gas phase concentration is low, because the gas metals condensed and was captured by the filter. The partitions in solid phase were raised when the operating time and sintering temperature increased. Under high temperature, more metals evaporated and escaped from the fly ash and they were collected in the exhaust gas.

When a water-washing treatment was applied, the metal partitions of Cr, Cd and Cu in the solid and gas phase were less than non-washing treatment. Small amounts of metal existed in the solid and gas phase could be the chlorine in the fly ash were washed out so metal chlorides were lightly vaporized, which represented that the water-washing was benefit for the sintering treatment. However, water-washing did not influence the reduction of the solid and gas phase of Pb. It was not clear and needs to be clarified that water-washing had different influences on the various metals.

3.5. Surface structure of the fly ash and sintering products

Fig. 5 depicts the SEM micrographs of porous and coarse surfaces of fly ash and sintering products. The fly ash exhibited the loose and cracked structure, as shown in Fig. 5(a). The surfaces of sintering products became smooth when the sintering temperatures rose to 900 °C, and there were no large pore structure on the surface. Under temperature of 900 °C, the phenomenon of liquid phase sintering occurred so the solidity became dense; in addition, the grain coalescence was observed also. The probable reason was that the sintering treatment aggregates the fracture of the fly ash under high temperature and the void of the fly ash was decreased. Fig. 6 shows the SEM of water-washing ash and sintering products. The structure of the washing ash is also loose and cracked which looks like that of the fly ash, as shown in Fig. 6(a). The sintering process for the washing ashes also compact them to a denser conditions so that the extractions of the metals lower.

3.6. XRD analysis

XRD analysis of fly ash, water-washing ash and sintering products is shown in Fig. 7. Fig. 7(a) shows that the main compositions of the fly ash are chlorides, the main compositions are $CaCl_2{\cdot}Ca(OH)_2{\cdot}H_2O$ and NaCl. In the existence of chlorine, the heavy metal chlorides are formed under sintering treatment. Because of their low boiling point, these metal chlorides vaporize to decrease the TCLP concentration of the sintering products. Besides, vaporization of the metal chlorides cause the sintering product of the fly ash loosen and less smooth than the sintering product of washing ash which will influence the leaching of the heavy metal inside of the fly ash. Water-washing treatment removed the chlorides so that the dominant composition switched to $Ca(OH)_2$, as shown in Fig. 7(b). Washout of chlorine was noticed to be helpful in the sintering treatment. Fig. 7(c) depicts the results of XRD analysis of the sintering products of fly ash. NaCl originally existed in the fly ash disappeared in the sintering product and new compositions such as Fe_2Si, Cd and $PbFe_2O_4$ were present. XRD result of sintered products of water-washing ash is shown in Fig. 7(d). Main compositions in the sintered water-washing ash were metal oxides of CaO, KO_2 and PbO_2 which have high boiling point. The compositions of Fe, Cd, Pb in the raw fly ash were washout in the water-washing ash and other metals reacted with the oxygen to form metal oxides of CaO, KO_2 and PbO_2 under sintering treatment.

4. Conclusion

The fly ash and bottom ash from the MSWI are valuable materials which can be reused. Besides, reuse of them not only can effectively prolong the life of the landfill but also reduce the risk of soil and underground water pollution if they are disposed. However, the fly ash is classified as the hazardous waste and cannot be reused directly because the TCLP leaching concentration of heavy metals exceeds the regulations. A useable process must be applied at first to handle the fly ash and decreases the TCLP concentration. The sintering treatment with a rotary kiln can be fitted to the incinerator and operated continuously. According to the experimental results, only the TCLP concentration of Pb in the fly ash was higher than the limitation. Sintering fly ash under temperature of 900 °C declined the concentration of Pb to agree the regulations. Water-washing treatment provided to be an effective process for decreasing the sintering temperature and time. Water-washing removed the chlorides of the fly ash to improve the quality of sintering products between 700

and 900 °C. The energy consumption of sintering declines if water-washing is used, however, the demand of the waste water treatment is deserved to be evaluated thoroughly.

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