



## Leaching characteristics of sulfates and chlorides from fly ash produced by municipal solid waste incineration

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### ABSTRACT

Fly ash (FA) produced by municipal solid waste incineration (MSWI) is a hazardous waste material that must be properly treated before disposal in landfills. The aim of the present study was to study the characteristics of MSWI-FA (i.e., morphology, particle size, mineralogical phase, and elemental content) before and after pretreatment, which is stabilization with the use of heavy metal chelating agents or the combination of stabilization and solidification with the use of cement. The results showed that pretreated MSWI-FA particles had more regular shapes with decreased diameters. Further, there was hardly any change to the mineralogical phase, which mainly consisted of calcium, aluminum, and silicon salts. The primary elements included O, Na, S, Cl, K, and Ca, and the weight percentage (wt%) and atomic percentage (At%) of Cl after pretreated was much higher than before. Metal leaching test of pretreated MSWI-FA shown that the concentrations of toxic metals in leachate were below the limits of standard, while the concentrations of alkaline metals, especially Na and K, were extremely high, ranging from 500 to 900 mg L<sup>-1</sup>. The results of a two-stage test of the leaching process revealed extremely high contents of the typical water-soluble salts in MSWI-FA. However, the pretreatment process failed to immobilize the sulfates and chlorides in MSWI-FA. In addition, as compared to chlorides, the sulfates in MSWI-FA transferred more quickly into the leachate, while the chlorides will gradually and may completely enter leachate. The development and application of chlorine- and sulfur-free reagents are recommended for the pretreatment of MSWI-FA, and a leachate collection system should be implemented for landfill disposal of MSWI-FA.

*Keywords:* Municipal solid waste; Landfill; Fly ash; Characteristics; Sulfates; Chlorides; Leaching test

### 1. Introduction

Municipal solid waste incineration (MSWI) is encouraged by the government of China because this process is relatively more efficient, less expensive, and requires less land use [1]. Fly ash (FA) produced by MSWI (MSWI-FA) is a hazardous waste material containing heavy metals and toxic dioxins that must be properly pretreated before disposal in landfills.

Previous studies have reported that though the constituents of MSWI FA were various by the time and city (or country), CaO, SiO<sub>2</sub>, MgO, Al<sub>2</sub>O<sub>3</sub> and alkali chlorides (mainly including KCl, NaCl, and MgCl<sub>2</sub>) can be found in most of MSWI FA samples [2,3], as well as various harmful heavy metals, such as Pb, Zn, Cd, Hg, Cu, and Ni [4,5]. Due to the high Ca content, pretreated MSWI-FA can be used as a binder in concrete [6–8]. The primary methods currently used for treatment (or pretreatment) of MSWI-FA

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include stabilization [9], solidification [10,11], thermal or hydrothermal treatment [12], and washing with water [13]. Most MSWI plants in China pretreat fly ash (FA) through stabilization with the use of heavy metal chelating agents or the combination of stabilization and solidification with the use of cement [14]. Prior to disposal in a landfill, pretreated MSWI-FA must meet the Standard for Pollution Control on the Landfill Site of Municipal Solid Waste of China [15]. Usually, most researchers concerned about the negative effects of heavy metals in MSWI-FA on the environment and human health during landfill disposal, even the environmental standard mentioned above only regulated the limit of leaching concentrations of heavy metal. However, the engineering issues arising from by the leaching of the water-soluble salts during the MSWI-FA landfill process seem to have been overlooked. The commonly used pretreated methods in MSWI plants hardly immobilize the water-soluble salts in MSWI-FA [16]. Moreover, many MSWI-FA landfills share the leachate treatment facility with MSW landfills. In consequence, the high concentrations of water-soluble salts inevitably enter into the leachate biochemical treatment system, which changes the composition of microorganisms in sludge and render the leachate collection and treatment systems unstable, and finally decreases the pollutants removal efficiencies [10,17,18].

The physical and chemical properties of MSWI-FA before and after different pretreatment process was investigated in this study. Moreover, the highlight of this work is to conduct the two-stage test of the leaching process to simulate the leaching characteristics of typical water-soluble salts (sulfates and chlorides) of pretreated MSWI-FA

following landfill disposal. The effects of sulfates and chlorides in MSWI-FA on leachate treatment systems of MSW landfills were identified in order to propose possible solutions.

## 2. Materials and methods

### 2.1. Samples

The MSWI-FA samples were collected from two MSWI plants (plants A and B) located along the Chengdu alluvial plain in China, which both used the same flue gas purification process. The acidified gas was removed with the dry/semi-dry method, while dust was captured with the use of a bag-type dust collector. MSWI-FA from the deacidification tower and bag-type dust collector was mixed in a storage tank. MSWI-FA samples from plant A were pretreated by stabilization with heavy metal chelating agents, while those from plant B were pretreated by stabilization and solidification. After pretreatment, the MSWI-FA samples were transported to a nearby sanitary landfill for safe disposal.

The MSWI-FA samples were respectively collected from the storage tanks and curing rooms of plants A and B (denoted as FA-A1/FA-B1 and FA-A2/FA-B2, respectively). Obviously, FA-A1 and FA-B1 was the MSWI-FA before pretreatment, while FA-A2/FA-B2 was the pretreated. In China, MSWI-FA is most commonly pretreated with stabilization or combination of stabilization and solidification, and the pretreatment methods were similar to that of plants A and B. The pretreatment methods and sampling points utilized in plants A and B are shown in Fig. 1.

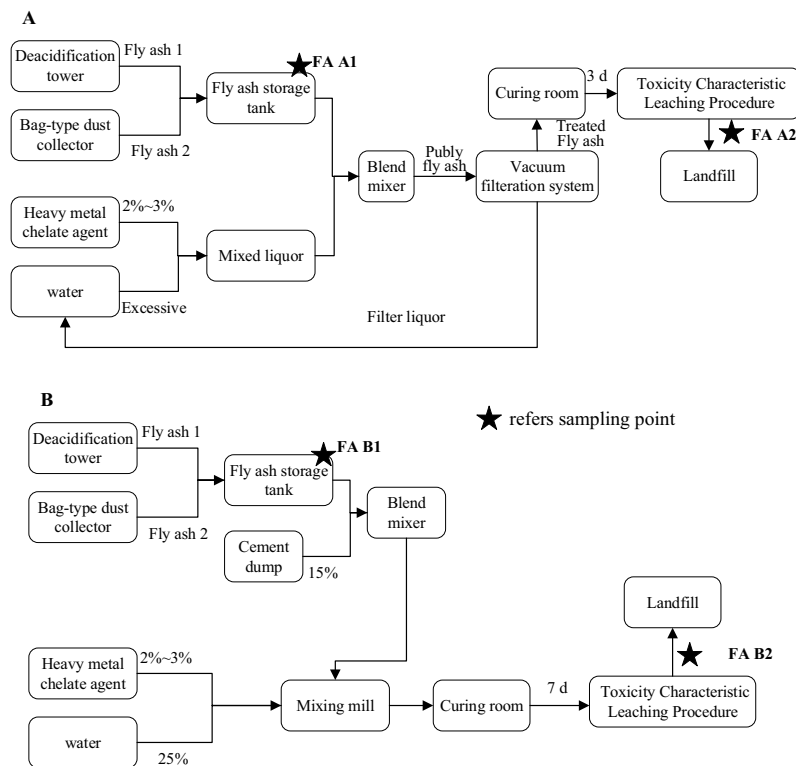


Fig. 1. MSWI-FA pretreatment processes and sampling points.

## 2.2. Methods

### 2.2.1. Analysis of sample characteristics

Particle size analysis was conducted using a dry laser particle size meter (HELOS-RODOS/M; Sympatec GmbH, Clausthal-Zellerfeld, Germany), while solid phase composition analysis was performed using an X-ray diffractometer (XD-2; Beijing PUXI General instrument Co. Ltd., Beijing, China) with the following parameters:  $2\theta$  ( $2\theta$ ) range,  $10^\circ$ – $70^\circ$ ; interval,  $0.02^\circ$ ; and scanning speed,  $4 \text{ min}^{-1}$ . Scanning electron microscopy (JSM 5900LV; JEOL, Ltd., Tokyo, Japan; working voltage, 20 kV; resolution, 30 nm) in conjunction with energy dispersive X-ray spectroscopy (SEM-EDS) was employed for analyses of the microscopic morphologies and elemental contents of the samples.

### 2.2.2. Leaching test

#### 2.2.2.1. Leaching test of metal

Chemical analysis was performed on the pretreated MSWI-FA (FA-A2 and FA-B2) to determine their heavy metal compositions. The leaching test procedure for Cu, Zn, Cd, Pb, Be, Ni, Cr and Ba carried out in accordance with identification standards for hazardous wastes-identification for extraction toxicity (Chinese Environmental Protection Administration, 2007) [19]. And the leaching test procedure for other metals (such as Hg, Se and Sn) was according to the solid waste-extraction procedure for leaching toxicity-acetic acid buffer solution method (Chinese Environmental Protection Administration, 2014) [20] and the concentrations of these metals were detected through inductively coupled plasma atomic emission spectrometer (ICP-AES, Agilent, 720ICP-OES).

#### 2.2.2.2. Leaching test of chlorides and sulfates

MSWI-FA contains high concentrations of alkali chlorides and sulphates, which may be leached out during landfill disposal and cause serious pollution to environment. The two-stage test of the leaching process was conducted to simulate the leaching characteristics of the sulfates and chlorides of pretreated MSWI-FA following landfill disposal. Briefly, 5 g of each sample was mixed with 25, 50, 75, or 100 mL of deionized water in a 100-mL conical flask, the liquid-to-solid ratios (L:S) of 5:1, 10:1, 15:1, or 20:1, respectively). Then, the conical flasks were fixed vertically to the oscillator, which was set to a frequency of 200 rpm for 1 h at  $25^\circ\text{C}$ . After oscillation, the supernatant was filtered through a  $0.45\text{-}\mu\text{m}$  filter membrane and the filtrate containing the

water-soluble substances was collected in a conical bottle, then dried the washed MSWI-FA samples prior to a second leaching process, during which the above steps were repeated. Afterward, the concentration of chlorides and sulfates in the filtrate were determined by ion chromatography (IC6000; Anhui Wan Yi Science and Technology Co., Ltd., Hefei, China).

## 3. Results and discussion

### 3.1. Sample characteristics before and after pretreatment

#### 3.1.1. Morphological analysis

The macro morphologies of the samples are shown in Fig. 2. Before pretreatment, the color of samples FA-A1 and FA-B1 was dark yellowish-brown with relatively large and closely distributed particles, which contained water and tended to coagulate.

After pretreatment, the color of samples FA-A2 and FA-B2 became pale gray and white, likely because of the addition of reagents during the pretreatment process. The particle size and pore structure of the pretreated samples were smaller than before treatment and the moisture contents had decreased.

SEM was used to analyze the microscopic morphologies of the samples to further illustrate morphological changes after pretreatment. SEM images of all four samples are shown in Fig. 3. The main microscopic morphological characteristics of sample FA-A1 were the formation of spherical bodies and some irregular crystals. After pretreatment, the particles of sample FA-A2 became globular or had a blocky floccule appearance, while the particle size had decreased, as did the space between particles, and the arrangement of particles was much denser. Before pretreatment, the microscopic morphologies of samples FA-B1 and FA-B2 were similar to the description above, while the particles were claviform or platy, with smaller diameters and more closely distributed after pretreatment.

Although the shapes of the particles were varied and irregular before pretreatment, the pretreated MSWI-FA samples were more regular in shape, with increased specific surface areas, consisting mainly of flocculent aggregates, and relatively more blocky and spherical bodies. The main purpose of the pretreatment process is the stabilization of heavy metals in order to minimize leaching during landfill disposal. The pretreated MSWI-FA contained cross-linked inclusions that were formed by chelating agents and heavy metals, as well as hydrated silicate crystals formed by cement hydration, which increased particle density after pretreatment.



Fig. 2. Macro-appearances of the samples.

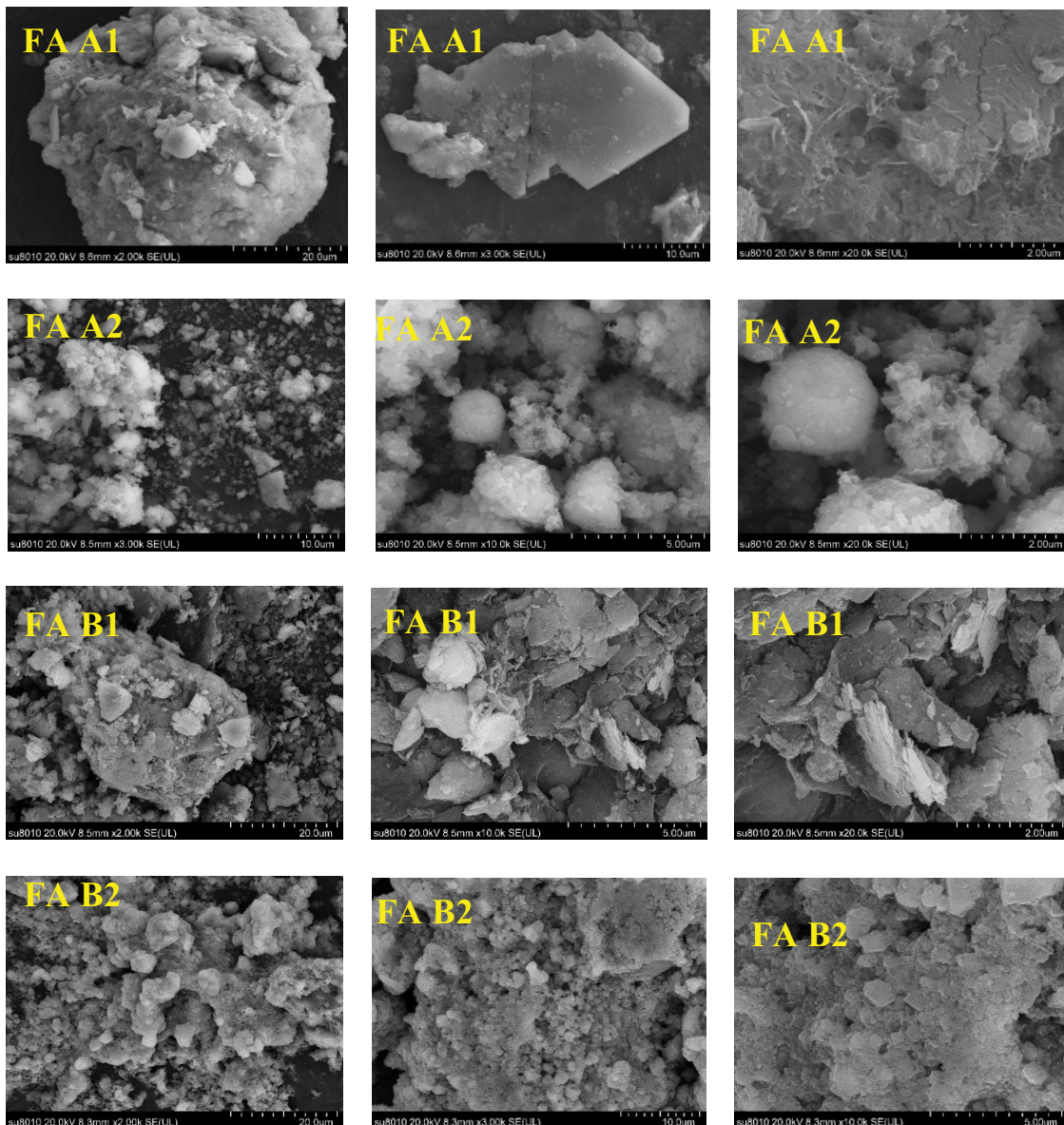


Fig. 3. SEM images of samples.

### 3.1.2. Analysis of particle size

As shown in Fig. 4a, before pretreatment, the FA-A1 particles with diameters of  $\geq 9.60 \mu\text{m}$  accounted for 10% of the cumulative volume of the total sample, that is,  $X_{10} = 9.6 \mu\text{m}$ . Similarly, particles with diameters of  $\geq 70.22$  and  $275.65 \mu\text{m}$  accounted for 50% and 90%, respectively ( $X_{50} = 70.22 \mu\text{m}$  and  $X_{90} = 275.65 \mu\text{m}$ ). Meanwhile, the Sauter mean diameter (SMD) and volume mean diameter (VMD) were  $22.79$  and  $109.27 \mu\text{m}$ , respectively. However, as shown in Fig. 4b, these values of FA-A2 were reduced after pretreatment (i.e.,  $X_{10} = 2.08 \mu\text{m}$ ,  $X_{50} = 14.29 \mu\text{m}$ ,  $X_{90} = 73.42 \mu\text{m}$ , SMD =  $6.76 \mu\text{m}$ , and VMD =  $28.53 \mu\text{m}$ ). Data of samples FA-B1 and FA-B2 are shown in Fig. 4c and d, respectively. The FA-B1 particle sizes were  $X_{10} = 2.97 \mu\text{m}$ ,  $X_{50} = 29.14$ , and  $X_{90} = 264.32 \mu\text{m}$  (SMD =  $9.80 \mu\text{m}$  and

VMD =  $86.26 \mu\text{m}$ ). For the pretreated samples, FA-B2, these data were  $X_{10} = 5.56 \mu\text{m}$ ,  $X_{50} = 31.79 \mu\text{m}$ , and  $X_{90} = 108.96 \mu\text{m}$  (SMD =  $13.62 \mu\text{m}$  and VMD =  $46.57 \mu\text{m}$ ).

Overall, the MSWI-FA particle sizes decreased after pretreatment. Notably, the  $X_{90}$  value decreased by at least 50%. These findings were consistent with the results of SEM analysis. According to particle size classification of sedimentary rocks, the particle sizes before treatment were similar to those of very fine sand ( $63\text{--}125 \mu\text{m}$ ) and medium-grade sand ( $>125 \mu\text{m}$ ), while the particle sizes after pretreatment were similar to those of coarse silt ( $16\text{--}63 \mu\text{m}$ ).

### 3.1.3. Mineralogical phase analysis

X-ray powder diffraction (XRD) was used to analyze the mineralogical phase components of the samples. Briefly,

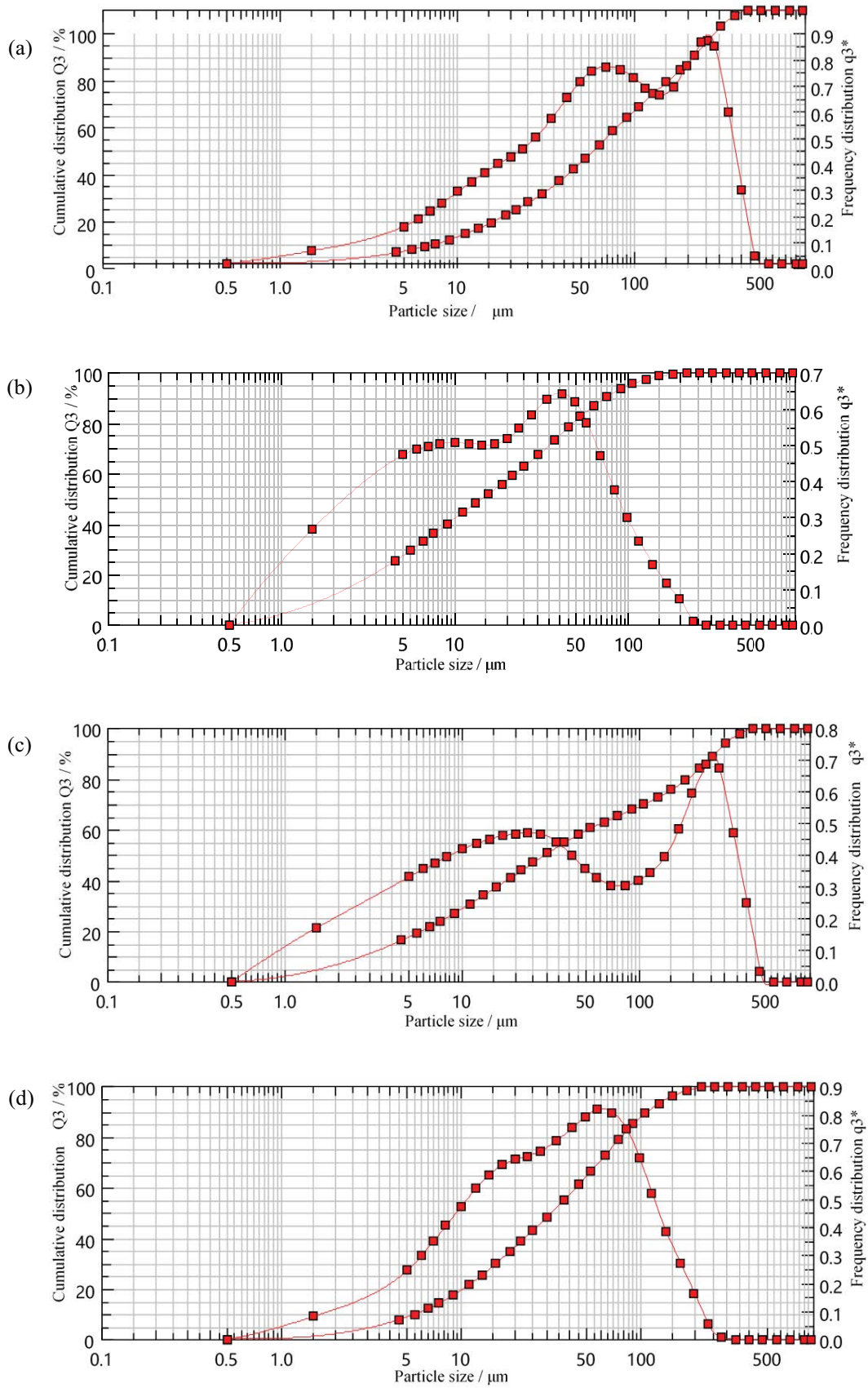


Fig. 4. Particle size analysis of samples (a) FA-A1, (b) FA-A2, (c) FA-B1, and (d) FA-B2.

5 g of each samples were ground into powder for analysis. As shown by the results presented in Fig. 5, the mineralogical phases of the MSWI-FA samples before and after pretreatment were similar. For samples FA-A1 and FA-A2 (Fig. 5a), there were peaks in the spectra at 18.1°, 28.5°, 29.9°, and 43.8°, indicating the presence of  $\text{Ca}(\text{ClO}_4)_2$  and  $\text{Ca}_{15}(\text{PO}_4)_6(\text{SiO}_4)_6$ , while diffraction peaks appeared at 21.0°, 25.6°, and 31.94°, indicating the existence of  $\text{Al}_2\text{O}_3$ . The spectra also revealed the existence of  $\text{CaO}$ ,  $\text{SiO}_2$ , and  $\text{FeS}_2$ . Samples FA-B1 and FA-B2 also contained  $\text{CaCO}_3$ ,  $\text{Ca}_2\text{S}_2\text{O}_7$ ,  $\text{NaCl}$  and  $\text{KCl}$  (Fig. 5b). Therefore, it can be concluded that stabilization or the combination of stabilization and solidification resulted in little change to the mineralogical phase of raw MSWI-FA, as the mineralogical phases were mainly calcium, aluminum, silicon salts, and water-soluble salts (e.g.,  $\text{NaCl}$ ). Of these, there were more types of calcium salts, which may have been due to the large amount of quick lime or alkali lime used for purification of flue gas.

### 3.1.4. Elemental content analysis

The results of elemental content analysis (EDS) of the samples are shown in Tables 1 and 2. The SEM-EDS images of samples could be found in supplementary information. The main constituent elements of the MSWI-FA samples before and after pretreatment were roughly the same (i.e., O, Na, S, Cl, K, and Ca).

For samples FA-A1 and FA-A2, the elements O, Cl, and Ca accounted for the highest weight percentage (Wt%) and atomic percentage (At%). Quick lime or alkali lime, which were used in the deacidification of flue gas, can react with acidic pollutants and transform into various calcium salts. The element O was mainly found in metal oxides, such as  $\text{CaO}$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ , and other materials identified by XRD analysis, while element Cl was mainly detected in the form of chlorides (i.e.,  $\text{KCl}$ ) or compound salts (i.e.,  $\text{Ca}(\text{ClO}_4)_2$ ). Elements O and Si were the top two constituents of sample FA-B1, while elements O, Cl, and Ca were the top three constituents of sample FA-B2. These results show that the main

Table 1  
Elemental content analysis of samples FA-A1 and FA-A2

Element	FA-A1		FA-A2		At%
	Wt%	At%	Element	Wt%	
O K	46.74	64.74	O K	42.48	61.57
Na K	8.44	8.13	Na K	6.30	6.35
Mg K	1.06	0.97	Mg K	0.83	0.79
Al K	0.32	0.26	Si K	1.19	0.98
Si K	1.35	1.06	S K	2.53	1.83
S K	3.68	2.55	Cl K	20.55	13.44
Cl K	14.80	9.25	K K	4.76	2.83
K K	3.24	1.83	Ca K	20.68	11.97
Ca K	20.01	11.06	Zn K	0.69	0.25
Fe K	0.37	0.15			
Total	100.00	100.00	Total	100.00	100.00

Table 2  
Elemental content analysis of samples FA-B1 and FA-B2

Element	FA-B1		FA-B2		At%
	Wt%	At%	Element	Wt%	
O K	58.94	73.18	O K	36.04	54.71
Na K	1.40	1.21	Na K	7.94	8.39
Mg K	0.41	0.33	Mg K	0.36	0.35
Al K	6.04	4.45	Si K	0.53	0.46
Si K	21.69	15.34	S K	2.21	1.67
S K	2.91	1.80	Cl K	28.59	19.58
Cl K	0.52	0.29	K K	5.25	3.26
K K	1.08	0.55	Ca K	19.08	11.56
Ca K	2.32	1.15			
Ti K	0.51	0.21			
Fe K	4.17	1.48			
Total	100.00	100.00	Total	100.00	100.00

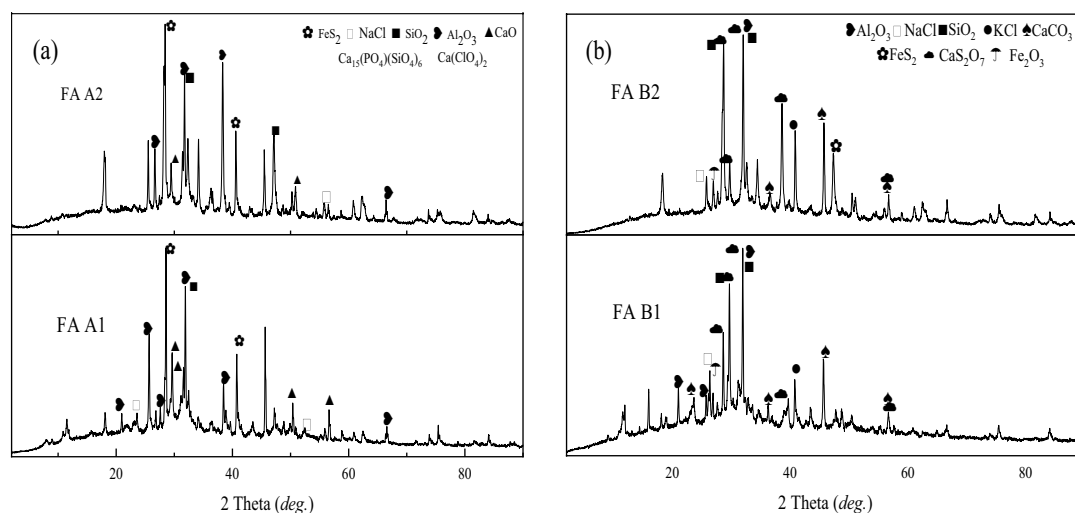


Fig. 5. XRD spectra of samples.

elements of the samples had slightly changed from before to after pretreatment, while the Cl content had increased.

### 3.2. Leaching test

#### 3.2.1. Metals leaching test

In order to investigate the capture effect of stabilization process on heavy metals in MSWI-FA, the metal leaching test was carried out on pretreated MSWI-FA samples (FA-A2 and FA-B2) and the results are shown in Table 3. The concentrations of heavy metals, involving Cu, Zn, Cr, Hg, Ni, As, Se, Pb, Be and Ba, in leachate were within the limits of the Chinese standard, indicating the MSWI-FA pretreatment process in these two MSWI plants was effective in stabilizing heavy metals [21,22]. However, we did not collect samples of leachate from the MSWI-FA landfill, so we could not fairly judge the actual situation of heavy metal leaching in the MSWI-FA landfill process.

The wt% and at% of typical alkaline metals (Na, K, Ca, Al) can be as high as 30% according to elemental content analysis. Therefore, their leaching concentration received attention in this work, and we found that their concentrations were much higher than that of toxic metals, especially the Na, the maximum concentration is about 900 mg L<sup>-1</sup>. Therefore, potassium salts and sodium salts are presumed to be the main components of the water-soluble salts in the leachate during MSWI-FA landfill disposal [23].

#### 3.2.2. Chlorides and sulfates leaching test

In the study of metal leaching behavior, we found that the leaching concentration of SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup> was extremely high, and which is consistent with previous studies [24–26].

Table 3

Metal leaching test on pretreated MSWI-FA and the MSW landfill disposal limits of China (Department of Environmental Protection of China, 2008)

Item	Concentration (mg L <sup>-1</sup> )		Landfill disposal limits (mg L <sup>-1</sup> )	
	FA-A2	FA-B2		
Heavy metals	Cu	N.D.	0.0036	40
	Zn	13.0	0.0191	100
	Cd	N.D.	0.0016	0.15
	Cr (VI)	N.D.	N.D.	1.5
	Total Cr	N.D.	0.0446	4.5
	Hg	N.D.	0.00053	0.05
	Ni	N.D.	0.0069	0.5
	As	0.00300	0.00312	0.3
	Se	N.D.	0.00412	0.1
	Pb	N.D.	0.112	0.25
	Be	N.D.	N.D.	0.02
	Ba	1.17	1.76	25
	Alkaline metals	Na	769.4	882.3
K		533.2	637.9	/
Ca		1.6	14.4	/
Al		33.4	26.7	/

Therefore, the concentrations of SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup> in supernatant after the different leaching processes were investigated and the results are shown in Fig. 6. In the figure C<sub>S1</sub> refers to the concentration of SO<sub>4</sub><sup>2-</sup> in supernatant after the first stage of the leaching test and C<sub>S2</sub> refers to the concentration of SO<sub>4</sub><sup>2-</sup> in supernatant after the second stage. C<sub>S2</sub> and C<sub>Cl2</sub> are defined similarly.

The L:S had the following effect on the concentrations of SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup> in supernatant (abbreviated as C<sub>S</sub> and C<sub>Cl</sub> respectively): 1) C<sub>S</sub> and C<sub>Cl</sub> were extremely high, and C<sub>S</sub> was almost higher than C<sub>Cl</sub> at the same L:S; 2) the L:S had a greater effect on C<sub>S</sub> as compared to C<sub>Cl</sub>; 3) the larger the L:S, the lower C<sub>S</sub> and the higher C<sub>Cl</sub>. As aforementioned, it can be inferred that the water-soluble salts may be mainly NaCl, KCl, Na<sub>2</sub>SO<sub>4</sub> and K<sub>2</sub>SO<sub>4</sub> are alkali and most are water-soluble. The concentrations of the sulfates and chlorides in MSWI-FA were extremely high [22,27,28], while the sulfates were more soluble than the chlorides. So, the C<sub>S</sub> was much higher, but notably decreased with a larger L:S. According to results of elemental content analysis, the Wt% and At% of Cl in samples were both relatively high, suggesting that the chloride content was relatively high in the samples. Furthermore, most were the chlorides, indicating that the chlorides in MSWI-FA will gradually and may completely transfer into the leachate with rainfall or surface runoff, although the sulfates transfer more quickly.

C<sub>S</sub> and C<sub>Cl</sub> in different leaching processes had the following characteristics: 1) C<sub>S1</sub> was greater than or close to C<sub>S2</sub>; 2) the smaller the L:S, the greater the difference between C<sub>S1</sub> and C<sub>S2</sub>; and 3) C<sub>Cl1</sub> was lower than C<sub>Cl2</sub>, but this difference was relatively small. These findings also demonstrate that the sulfates can transfer into leachate more quickly.

In addition, the C<sub>S</sub> and C<sub>Cl</sub> after pretreatment (FA-A2 and FA-B2) were greater than before pretreatment (FA-A1 and FA-B1), indicating that the pretreatment processes (stabilization and the combination of stabilization and solidification mentioned in section 2.1) in this study were impossible to immobilize the sulfates and chlorides, and even increase the contents of the typical water-soluble salts in MSWI-FA [29]. As a possible explanation for this finding, the reagents used in the pretreatment process (e.g., heavy metal chelate agents) may contain many water-soluble salts that can easily wash out. It has to be noted that in the pretreatment process of fly ash in MSWI plant B, only a low dose of cement was added (the mass ratio of cement to fly ash was 1%), the physical barrier of cement to sulfate was not obvious [30,31].

In China, most landfills that accept MSWI-FA for disposal do not have a separate leachate collection system, but share a system with the municipal landfill. Furthermore, standardization of the daily management of most landfills is lacking, especially in regard to daily cover. So, the typical water-soluble salts are easily transferred into the leachate, resulting in an increase in the soluble conductivity of leachate, which has a negative impact on the treatment of leachate [32]. Therefore, the following recommendations are proposed, (1) chlorine- and sulfur-free reagents should be developed and applied during the pretreatment process. (2) Landfills should have separate leachate collection systems for MSWI-FA, (3) management practices should be standardized to minimize the transfer of sulfates and

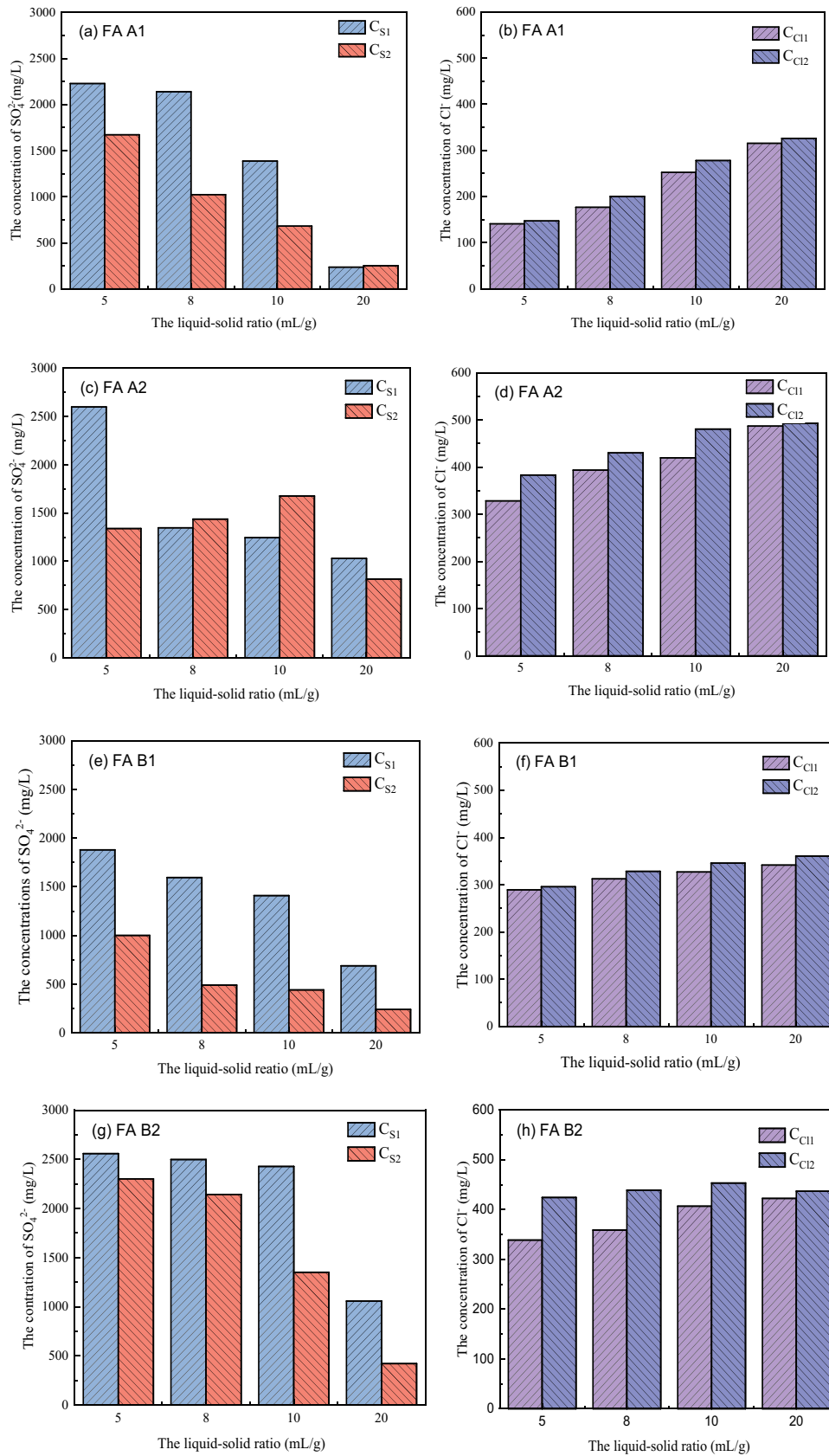


Fig. 6. SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup> concentrations in the supernatants of samples after different leaching processes (a) and (b) FA-A1, (c) and (d) FA-A2, (e) and (f) FA-B1, and (g) and (h) FA-B2.



chlorides into leachate. And (4) for MSWI-FA landfill areas that share leachate treatment systems with MSW landfills, it is recommended that control the salinity of leachate from MSWI-FA landfill area be less than 0.4% in consideration of microbial growth needs [33]; meanwhile, the salt-tolerate bacteria, such as the sulfate-reducing bacteria after domestication should be added into the biological treatment unit to reduce the impact of high-salt leachate on the influent water quality [34].

#### 4. Conclusions

The shapes of MSWI-FA particles were varied and irregular before pretreatment. After pretreatment, the particles become more regular in shape, with decreased particle sizes and increased specific surface areas, mainly in the form of flocculent aggregates with relatively more blocky and spherical bodies. Stabilization or the combination of stabilization and solidification caused little change to the mineralogical phase of raw MSWI-FA. The mineralogical phase mainly consisted of calcium, aluminum, and silicon salts, while the main elements consisted of O, Na, S, Cl, K, and Ca.

The concentrations of toxic metals in leachate were below the limits of standard, while the concentrations of alkaline metals, especially Na and K, were extremely high, ranging from 500 to 900 mg L<sup>-1</sup>. The concentrations of sulfates and chlorides in MSWI-FA were extremely high. It is impossible to immobilize sulfates and chlorides in MSWI-FA, and even increase the contents of the water-soluble salts by stabilization or the combination of stabilization and solidification. The typical water-soluble salts will transfer from MSWI-FA into leachate more quickly, while water-soluble chlorides will gradually and may completely enter leachate. We recommend the development and application of chlorine- and sulfur-free reagents during the MSWI-FA pretreatment process, and the construction of separate leachate collection systems for landfills.

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