RESEARCH ARTICLE

Effect of alkaline washing treatment on leaching behavior of municipal solid waste incineration bottom ash

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Abstract

This study aimed to find an effective, inexpensive, and safe washing treatment for municipal solid waste incineration bottom ash (MSWIBA) in order to reduce its potential harmful effects in disposal and recycling. The washing solutions, namely tap water (TW), saturated lime water (SLW), and wastewater from concrete batching plant (WW) were used to wash MSWIBA at different liquid–solid (L/S) ratios and for different durations. Leaching behavior of some heavy metals, chloride, and sulfate from MSWIBA was tested and evaluated. From the TCLP leaching test, when the L/S ratio was above 5, WW was the most effective solution in reducing As, Cd, Se, and Sb emissions from MSWIBA. The calcium and iron ions present in the WW were essential for controlling the leaching of As, Cd, and Sb from MSWIBA due to the formation of stable crystalline pharmacosiderite, cadmium hydroxide sulfate, and hydromeite during the washing process. Using WW showed the best effect in removing sulfate from MSWIBA. At a L/S ratio of 10, about 83% of the sulfate could be removed from MSWIBA after 20 min of washing. The L/S ratio was most influential in removing chloride from MSWIBA. The three washing treatments chosen were effective in reducing the chloride level in MSWIBA to below the level of hazardous waste. Nevertheless, there were still substantial amounts of chloride remaining in the treated MSWIBA. Under the Dutch Building Materials Decree, the treated MSWIBA may be used as a building material in parts which allow isolation, control, and monitoring (ICM).

Introduction

Incineration of municipal solid waste (MSW) is becoming more common because of various benefits such as great reduction in waste volume and conversion of waste to energy. During the incineration process, three types of solid residues are produced, including bottom ash (MSWIBA), fly ash, and air pollution control residue. Over the years, many techniques have been developed for the pre-treatment of incineration residues to reduce subsequent leaching of pollutants and harmful substances to environment and product masses respectively (Fan et al. 2021; Lei et al. 2021). Typical pre-treatment methods can be generally classified into extraction method and stabilization method. Extraction is a way to reduce the pollution potential of residues by removing pollutants in advance. In applying pre-treatments of this kind, consideration should be given not only to efficiency in removing pollutants, but also to the economical use of water, chemicals, and energy. Common extraction methods include water washing, alkaline washing, acid leaching, and extraction with chelating agents (Leštan et al. 2008; Tandy et al. 2004). Novel methods include electrodialysis and supercritical fluid extraction (Couto et al. 2021; Isci and Kaltschmitt 2021). On the other hand, stabilization method is through stabilization of contaminants to control the potential pollution problem from wastes (Lu et al. 2021; Kearns et al. 2021). The most common stabilization method is by binding with cement (Conner and Hoeffner 1998; Guo et al. 2020).

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Other stabilization methods include thermal stabilization, carbonation/weathering, and stabilization with chemical agents (Zhu et al. 2021).

MSWIBA is in general not regarded as a hazardous waste based on many research findings (Cioffi et al. 2011; Schnabel et al. 2021), and it has a potential for reuse as construction minerals to substitute some natural granular materials in geotechnical works or in concrete (Qiao et al. 2008; Lynn and Dhir 2017). However, MSWIBA contains various harmful components such as heavy metals and soluble salts which may threaten human health or natural environment when they leach out. It is therefore highly desirable to treat MSWIBA to reduce possible leaching of such elements before recycling MSWIBA in construction works. Leaching is a process of transporting mass from a solid material to the surrounding liquid phase. Conditions affecting leaching include pH, ionic strength, buffer capacity, redox, liquid-to-solid (L/S) ratio, solvents, and the presence of complexing agents (Roberts 2018; Šljivić-Ivanović and Smičiklas 2020).

Due to the relatively low contents of heavy metals in MSWIBA, acid leaching and the use of chelating agents need not be applied for general removal of heavy metals to save money. Normally, washing with water is adopted for removing soluble chloride and sulfate in MSWIBA together with a little amount of heavy metals inside a quench tank or soil cleansing facility in practice. In this study, attention was paid to four elements, namely arsenic (As), cadmium (Cd), selenium (Se), and antimony (Sb). As, Cd, and Se are volatile elements, which can easily condense on the surface of fine ash particles during the combustion of MSW. Very stringent limits have been set on the leaching of these metals from wastes due to their potential carcinogenic propensities. In general, the extent of leaching of a heavy metal from a particle is largely affected by the surface solubility of the metal which varies with the speciation of the metal and the leaching conditions around the particle. Different species of a metal often have different solubilities while leaching conditions such as pH and redox potential also control their solubility. Other relevant leaching conditions include L/S ratio, solvents used, and the intensity of the contact. The means to reduce the pollution potential of MSWIBA can either by removal of pollutants through some form of leaching in advance or in the opposite way by stabilizing pollutants in MSWIBA. For pollutant stabilization, lime/calcium hydroxide can be used to precipitate some heavy metals into their insoluble hydroxide forms in an alkaline environment (Charerntanyarak 1999; Tünay 2003). In other words, lime/calcium hydroxide is a precipiting agent in a high pH condition.

As chloride is highly soluble in water, Kim et al. (2021) showed that with L/S ratio of 2.5 and just 5 min of washing, 77% of the chloride could be removed from a mixture of incineration bottom and fly ashes. Although different sulfate

minerals have different solubility, the relatively high solubility of sulfate in MSWIBA was noted in many studies (Belevi et al. 1992; Stegemann et al. 1995). The successful use of a soil cleansing facility to remove sulfate salts from MSWIBA by washing was reported by Boddum and Skaarup (2000). Contrary to acid washing which may promote the dissolution of most heavy metals in ashes/residues, it is believed that an alkaline solution may demobilize some heavy metals in MSWIBA (Jang et al. 2005; Cao et al. 2019), thereby reducing its pollution potential while maintaining efficiency in removing chloride and sulfate.

Although the use of tap water (TW) is the most common washing treatment method, it would be very worthwhile to investigate the influence of alkaline washing treatment on the leaching behavior of MSWIBA in depth, in particular using industrial alkaline waste water as a washing media. In this research, three washing solutions were studied for comparison of performance. The effectiveness of TW and two alkaline solutions, namely saturated lime water (SLW) and reclaimed waste water (WW), from a local ready-mix concrete batching plant in treating MSWIBA was evaluated. Wastewater in ready-mix concrete batching plants is largely generated from washing concrete trucks and equipment, which is a highly alkaline and calcium-rich solution (Monkman et al. 2021; Sojobi et al. 2021). The main objective of this fundamental study is to examine the feasibility of applying a one-step washing method using alkaline solution to treat the contaminants in MSWIBA. This study provides new insights into the potential of washing treatment in removing heavy metals, chloride, and sulfate constituents in MSWIBA.

Materials and methods

MSWIBA

MSWIBA was collected from an incinerator in Hangzhou, China, and then stored in a laboratory also in China for over 1 year. Table Table 2 Leaching of heavy metals from MSWIBA as received Please provide the significance of the values in bold found in Table 2. MetalUS TCLP leaching test (mg/L)EN leaching test (mg/kg) LeachingLimits (TCLP limits, RCRA metals TCLP analysis) LeachingLimits ("European Council Decision 2003/33/CE 2003) Inert waste Non-hazardous wasteAs0.22050.1500.52Ba0.1321001.82220100Cd1.90010.0490.041Cr < 0.00150.0140.510Cu2.187-0.154250Ni0.296-0.0370.410Pb < 0.0150.0340.510Sb0.350-0.0510.060.7Se0.40010.0630.10.5Zn24.459-0.3164501 presents the oxide compositions and physical properties of the MSWIBA. It is clear from Table 1 that the raw MSWIBA contained many different heavy metals. Table 2 gives the metal

Table 1 Chemical and physical properties of MSWIBA as received

Oxide (wt. %)			Metal (mg/kg)	
Na$_2$O	3.14		As	21.82
MgO	2.51		Ba	1170.53
Al$_2$O$_3$	10.83		Cd	78.55
SiO$_2$	37.65		Cr	323.00
Fe$_2$O$_3$	6.63		Cu	1638.53
P$_2$O$_5$	3.52		Ni	111.71
TiO$_2$	1.27		Pb	682.12
K$_2$O	1.77		Sb	3.37
CaO	24.64		Se	4.69
SO$_3$	4.55		Zn	6701.93
LOI	2.52			
Physical properties	Particle size (mm)			0.15~2.36
	Blaine fineness (m^2/kg)			2356.3
	Water absorption (%)			15.39
	Saturated surface dry density (g/cm^3)			1.32
	Oven-dry density (g/cm^3)			1.13

Table 2 Leaching of heavy metals from MSWIBA as received

Metal	US TCLP leaching test (mg/L)		EN leaching test (mg/kg)		
	Leaching	Limits (TCLP limits, RCRA metals TCLP analysis)	Leaching	Limits (EuropeanCouncil Decision 2003/33/CE 2003)	
				Inert waste	Non-hazardous waste
As	**0.220±0.006**	**5**	**0.150±0.003**	**0.5**	**2**
Ba	0.132	100	1.822	20	100
Cd	**1.900±0.006**	**1**	**0.049±0.002**	**0.04**	**1**
Cr	<0.001	5	0.014	0.5	10
Cu	2.187	-	0.154	2	50
Ni	0.296	-	0.037	0.4	10
Pb	<0.01	5	0.034	0.5	10
Sb	**0.350±0.005**	**-**	**0.051±0.002**	**0.06**	**0.7**
Se	**0.400±0.007**	**1**	**0.063±0.004**	**0.1**	**0.5**
Zn	24.459	-	0.316	4	50

concentrations leached from the MSWIBA under the US TCLP leaching test (EPA Method 1311, 1992) and the EN leaching test (EN 12457-2 2002), respectively. Leaching concentrations from the US TCLP leaching test were checked against the disposal criteria TCLP limits, RCRA metals TCLP analysis. The leachable amount of Cd was found to be higher than the allowable limit of 1 mg/L. The results of the EN leaching test were compared with the leaching limit values for disposal of inert and non-hazardous materials at landfill sites ("European Council Decision

2003/33/CE 2003). It was found that only Cd released from the MSWIBA exceeded the limit value for storage of inert waste. The leaching concentrations of Se and Sb from the MSWIBA were close to their threshold limits. The leaching of toxic metals Cd, Se, and Sb from the MSWIBA should therefore be effectively lowered. The leaching of strong acid anions was also checked against the limits stated in the waste acceptance criteria under the European Council Decision 2003/33/EC (2003). The fluoride ion concentration measured by the ion chromatography was 3.5 mg/kg which fell below the threshold limit of 10 mg/kg for inert waste. However, the chloride and sulfate concentrations were 64,805 and 12,150 mg/kg respectively, which were far above the limit values for acceptance of waste in inert landfills, i.e., 800 and 1000 mg/kg respectively.

Washing treatment

Although a higher L/S ratio should likely give a better result in stabilizing and/or removing contaminants, benefits and cost have to be balanced to optimize the treatment process. Therefore, this study investigated the performance of washing with three L/S ratios (2.5, 5, and 10) and three washing durations (10, 20, and 30 min), which had been commonly adopted in similar studies (Wang et al. 2016; Li et al. 2021).

Tap water (TW)

TW was potable water supplied by the water authority to the general public. Its pH was 7.80 ± 0.23. This study evaluated the performance of TW washing with three L/S ratios and three washing durations which provided baselines for comparing the performance of the alkaline washing solutions (SLW and WW).

Saturated lime water (SLW)

SLW was prepared in the laboratory by adding calcium hydroxide pellets in distilled water until saturation. The solubility of calcium hydroxide was around 1.7 g/L. There were several reasons for adopting SLW for washing. Firstly, the pH value of SLW was 11.41 ± 0.37, which was close to that of wastewater (WW) thus facilitating comparison of the performance of the two solutions. Secondly, SLW was free of contaminants. Moreover, SLW could be prepared conveniently.

Wastewater (WW) from ready-mixed concrete batching plant

WW was obtained from a major ready-mixed concrete production plant in Hong Kong, which generated a large amount of WW from washing concrete leftover in equipment

and trucks. Various impurities of cationic elements such as Ca^{2+} (453.0 ± 7.0 ppm), Na^+ (117.5 ± 1.0 ppm), K^+ (12.4 ± 0.1 ppm), and Fe^{3+} (10.1 ± 3.2 ppm) existed in the WW. The pH value of WW was 11.58 ± 0.52. Under a high pH environment, some heavy metals may be demobilized more readily. Reclaiming WW for controlling the pollution potential of MSWIBA, if feasible, would be environmental friendly and cost effective.

Washing process of MSWIBA

Twenty-seven sets of MSWIBA samples were prepared for different washing treatments, as listed in Table 3. Washing was carried out in a set-up which allowed stirring the mixture at a speed of 40 r/min to ensure high contact intensity during the washing period. MSWIBA samples having washed with TW, SLW, or WW are denoted as MSWIBA$_{TW}$, MSWIBA$_{SLW}$, or MSWIBA$_{WW}$ respectively. The detailed washing procedures are as follows:

(1) Prepare the appropriate amount of washing solution according to the prescribed L/S ratio
(2) Wash the MSWIBA inside the mixer for the prescribed duration
(3) After washing, separate the MSWIBA from the fluid with a sieve of 150 μm The reason for choosing 150 μm sieve was to avoid unduly long time for separating the washing solution and the MSWIBA.
(4) Oven dry the samples at 105 °C for 24 h
(5) Grind the ash particles to sizes of less than 2.36 mm
(6) Carry out experimental tests on individual samples
(7) Compare the influences of different washing treatments on the leaching behavior of residual samples. The results given are the averages of three measurements.

Testing methods

The oxide compositions were determined using the X-ray fluorescence (XRF) technique. The total metal contents were determined with aqua regia digestion. The particle size distribution was detected using a laser diffraction particle size analyzer. The Blaine fineness was determined according to ASTM C204-18e1 (2018) (ASTM C204-18, 2019). The particle densities and water absorption value were measured as per ASTM C127 (ASTM C127-15, 2015) and C128 (ASTM C128-15, 2015), respectively. The chloride concentration was measured following the procedure of ASTM C1152 (2012). The sulfate concentration was measured by a SulfaVer 4 turbidimetric kit developed by the HACH company based on HACH Method 8051 (2003), which is equivalent to US EPA Method 375.4. The US toxicity characteristic leaching procedure (TCLP) follows the EPA Method 1311 (1992) while the EN leaching testing procedure follows the EN 12457-2 (2002). Metal concentrations measured on analytes passing through 0.45 m filters were determined by the inductively coupled plasma-atomic emission spectrometry (ICP-AES, PerkinElmer Optima 3300 DV). The crystalline phases were analyzed by the X-ray powder diffraction (XRD) technique.

Results

Chloride concentrations in the treated MSWIBA

The chloride concentration in the raw MSWIBA was 64,805 mg/kg. Figure 1 shows the influences of various washing conditions, namely washing solutions (or fluid alkalinity), L/S ratios, and washing time, on the concentrations of chloride that leached out. In general, the fluid alkalinity and washing time of the fluid did not have significant effects on the effectiveness of the chloride removal. Higher L/S ratios in the washing process greatly enhanced the removal of chloride in the MSWIBA as evident from the notable reductions in the chloride concentrations. In fact, a low L/S ratio of 2.5 was sufficient to reduce chloride level to below the hazardous waste limit of 25,000 mg/kg ("European Council Decision 2003/33/CE 2003). After different washing treatments applied in this study, concentrations of the chloride were lower than that of the raw MSWIBA. However, the chloride concentrations in the treated MSWIBA were still much higher than the inert waste limit of 800 mg/kg specified in European Council Decision 2003/33/EC (2003).

Table 3 Combination of washing variables

| Duration (min) | Washing fluid | | | | | | | | |
L/S	TW			SLW			WW		
2.5	10	20	30	10	20	30	10	20	30
5	10	20	30	10	20	30	10	20	30
10	10	20	30	10	20	30	10	20	30

Fig. 1 Chloride concentrations of treated MSWIBA
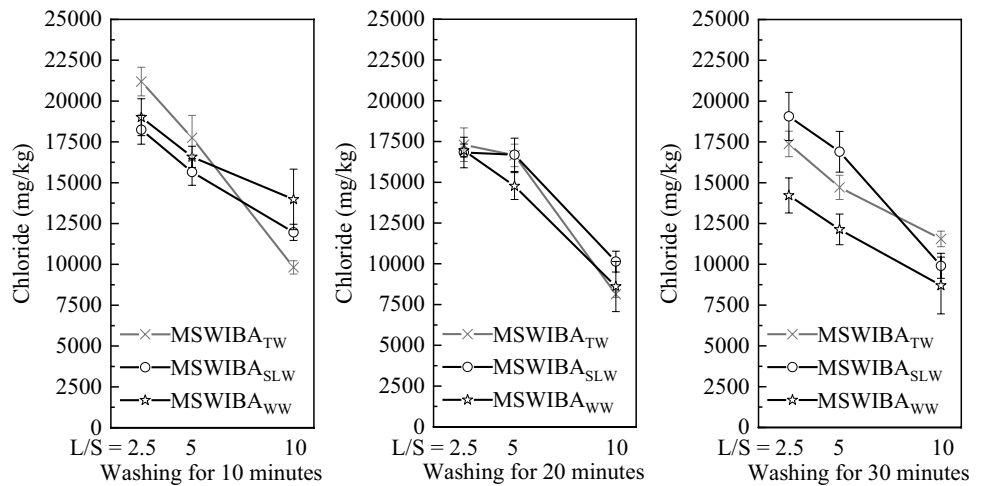
Fig. 2 Sulfate concentrations of treated MSWIBA
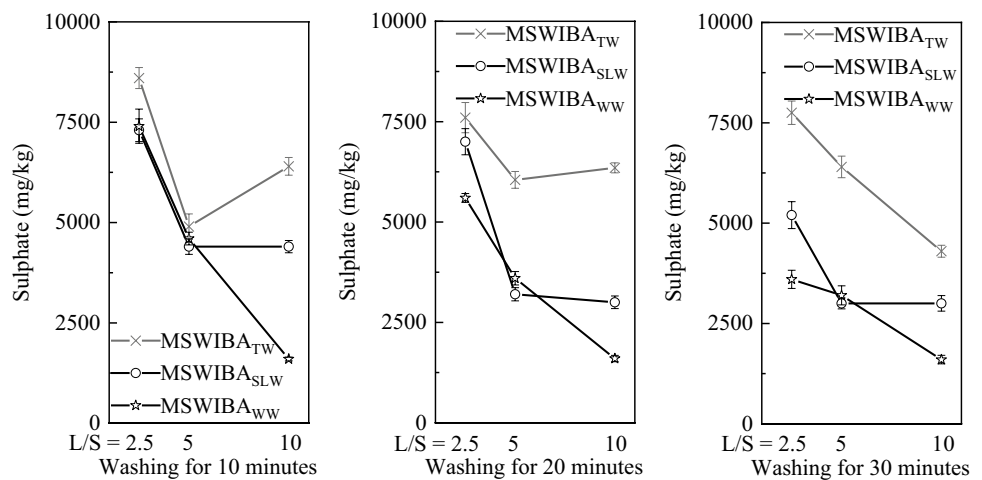
Sulfate concentrations in the treated MSWIBA

The sulfate concentration in the raw MSWIBA was 12,150 mg/kg. Figure 2 shows the sulfate concentrations of the treated MSWIBA samples. It can be seen that in general, WW was the most effective in removing sulfate in the MSWIBA, followed by SLW, implying that high alkalinity was conducive to the removal of sulfate. Besides, the rather limited effects from increasing L/S ratios and washing time are also notable, indicating that solubilization of sulfate in the MSWIBA was less easy compared with chloride. Nevertheless, the sulfate removal efficiency reached over 85% with WW washing at the high L/S ratio of 10. Comparing with the sulfate limit specified in European Council Decision 2003/33/EC (2003), the sulfate concentration in the treated MSWIBA was still above the limit value of inert waste (1000 mg/kg).

Metal leaching characteristics of the treated MSWIBA

pH values of the filtered analytes

MSWIBA samples after washing treatments were prepared for the leaching tests in order to compare their leaching characteristics to that of the raw MSWIBA. Leaching tests were conducted according to TCLP (EPA Method 1311, 1992) under the acidic condition and EN 12457-22 (2002) with DI water. The pH values of the filtered analytes were then determined using a pH meter, and the metal concentrations in the filtered analytes were measured using the ICP equipment. Table 4 shows the pH values of the filtered analytes from the TW-, SLW-, and WW-treated MSWIBA samples compared with the raw MSWIBA sample. The pH value of the filtered analyte from the raw MSWIBA sample under the EN

Table 4 pH values of filtered analytes

		US TCLP leaching test					EN leaching test			
		Raw	Treated MSWIBA			Raw		Treated MSWIBA		
		6.6	L/S = 2.5			9.4		L/S = 2.5		
			10 min	20 min	30 min			10 min	20 min	30 min
	TW		7.6 (±0.2)	7.0 (±0.2)	7.1 (±0.1)		TW	9.8 (±0.5)	10.2 (±0.1)	10.1 (±0.1)
	SLW		7.6 (±0.2)	6.5 (±0.3)	6.8 (±0.1)		SLW	10.6 (±0.1)	10.1 (±0.5)	10.5 (±0.1)
	WW		7.1 (±0.2)	6.6 (±0.2)	6.5 (±0.3)		WW	10.3 (±0.4)	10.6 (±0.1)	10.5 (±0.3)
			L/S = 5					L/S = 5		
			10 min	20 min	30 min			10 min	20 min	30 min
	TW		7.6 (±0.2)	7.1 (±0.1)	7.1 (±0.2)		TW	10.1 (±0.4)	10.1 (±0.3)	10.4 (±0.1)
	SLW		7.3 (±0.1)	6.6 (±0.2)	6.6 (±0.1)		SLW	10.3 (±0.5)	10.6 (±0.2)	10.9 (±0.1)
	WW		6.9 (±0.1)	6.8 (±0.1)	6.6 (±0.2)		WW	10.7 (±0.1)	10.7 (±0.1)	10.3 (±0.5)
			L/S = 10					L/S = 10		
			10 min	20 min	30 min			10 min	20 min	30 min
	TW		7.3 (±0.1)	7.1 (±0.1)	7.0 (±0.2)		TW	10.4 (±0.1)	9.8 (±0.5)	10.4 (±0.1)
	SLW		7.0 (±0.2)	6.5 (±0.2)	6.6 (±0.1)		SLW	10.6 (±0.1)	10.7 (±0.1)	10.6 (±0.1)
	WW		6.8 (±0.1)	6.7 (±0.1)	6.7 (±0.2)		WW	10.8 (±0.1)	10.8 (±0.1)	10.9 (±0.1)

leaching test was 9.38 ± 0.36. Based on the EN leaching test, the filtered analytes from the TW, SLW, and WW washed MSWIBA samples exhibited higher alkalinity compared with the raw MSWIBA sample. The increments ranged from approximately 4 to 15%. During the washing process, some cationic ions present in the washing solutions were absorbed into the porous MSWIBA particles and attached to the pore surfaces of the washed MSWIBA particles (Xuan et al. 2019; Sun et al. 2021). In the subsequent 24 h DI water leaching, alkaline components leached from the samples, resulting in higher pH values in the leachates. The results show less influence from washing time and L/S ratios on the pH values of filtered analytes from TW, SLW, and WW treated MSWIBA, respectively, considering the standard deviation as a precision measure of experiments. The pH values of filtered analytes show a notable change with different washing fluids due to their different alkalinity. From the results, higher alkalinity of the washing fluids resulted in higher pH values of the filtered analytes. As SLW and WW had higher alkalinity, little amounts of alkaline components might be retained in the samples after the washing treatments, leading to higher alkalinity of their leachates generated.

The pH value of the filtered analyte from the raw MSWIBA sample under the US TCLP leaching test was 6.63 ± 0.23. It can be seen from Table 4 that samples washed with TW exhibited higher pH than that of the raw sample. The leachates from samples having washed for 10 min exhibited higher pH values than those washed for a longer time (20 ∼ 30 min). In addition, with 10 min, the pH values of leachates decreased with increasing L/S ratios, but with a longer washing time (20 ∼ 30 min), L/S ratio had less effect on the pH. As most soluble chlorides and sulphates had been removed from the ash during the washing pretreatment, the analyte solutions from the subsequent leaching tests contained less acidic components resulting in higher pH compared with the untreated ash. The pH values of TW treated MSWIBA leachates were generally higher compared with SLW and WW treatment, indicating that the use of a stronger alkaline washing solution led to a reduction in the pH value of the leachate from the treated sample. This phenomenon is contrary to the results from the EN leaching test, in which the leachate pH increased with the alkalinity of the washing fluid. This phenomenon can be explained by the difference in the alkali ion contents in the MSWIBA residues after washing with different fluids. When the concentrations of alkali reactants were high, the chemical equilibria favored the forward reaction which transformed from left to right leading to reduction in the solution pH when the same amount of acid was added to the material.

Metal leaching from the treated MSWIBA

Figures 3, 4, 5, and 6 show the leaching concentrations of heavy metals from MSWIBA after different washing treatments. Figure 3 shows the As concentrations in leachates after treatments and the changes of concentrations compared with the untreated samples. Data variables and sudden changes in the EN leaching test results make it difficult to compare effectiveness of different washing treatments. It is interesting to reveal from the US TCLP leaching test that WW was highly effective in removing As from MSWIBA. The results showed that the removal efficiency increased noticeably by washing with a high L/S ratio. As leached in great concentrations under the tests when the L/S was 2.5, the concentration significantly decreased with increasing L/S ratios.
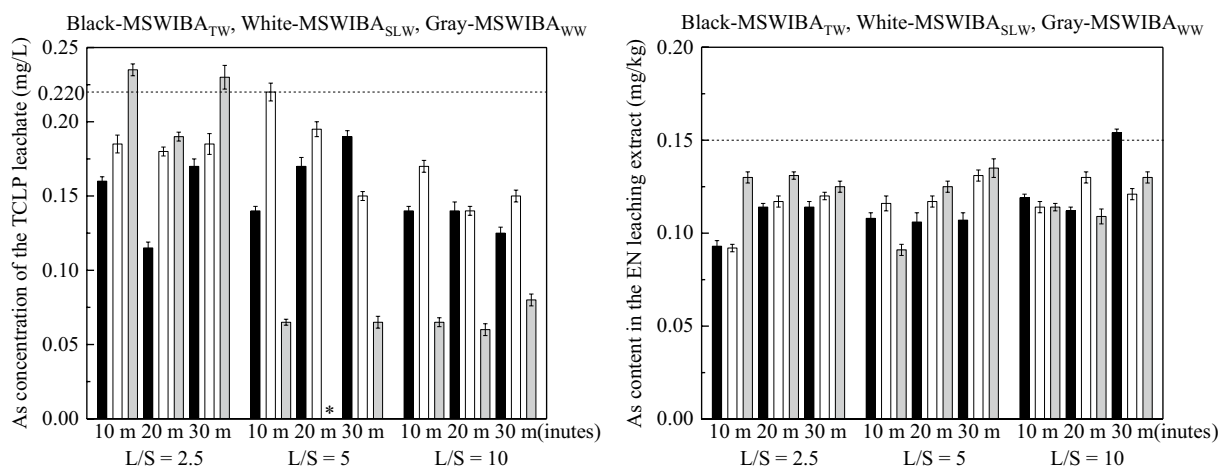
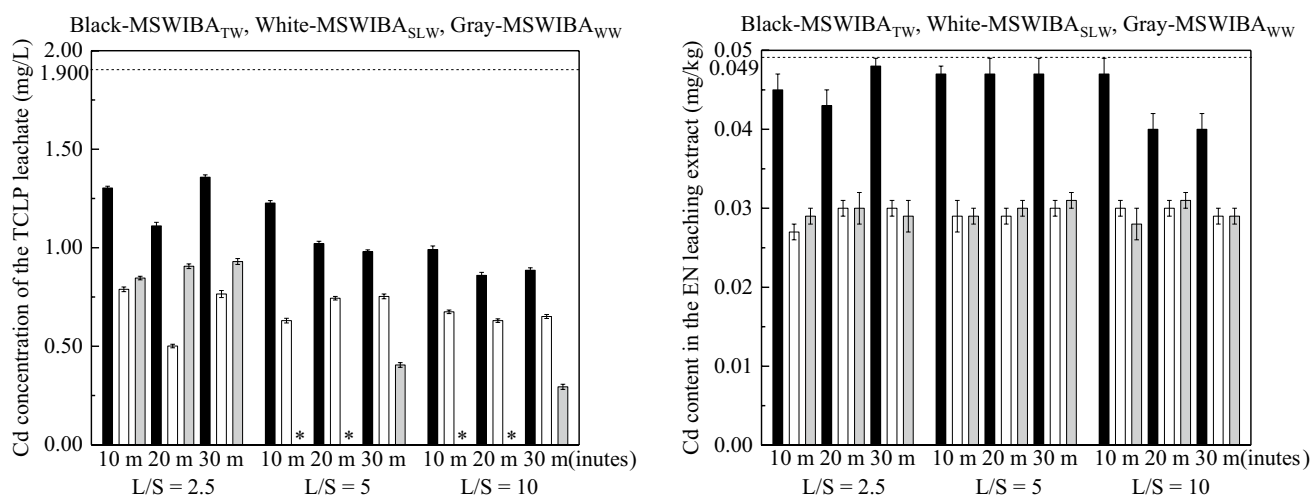**Fig. 3** As leaching concentrations (*below detection limit)

Fig. 4 Cd leaching concentrations (*below detection limit)

Figure 4 shows the leaching concentrations of Cd measured in leachates from both testing procedures. As evident from the results, alkaline washing treatment is a very effective way to alleviate the leaching risk of Cd to the environment. Cd leaching from the MSWIBA samples after various washing treatments with SLW and WW fell below waste acceptance criteria for disposal to landfill set by the US Environmental Protection Agency, i.e., 1 mg/L, or the European Union, i.e., 0.04 mg/kg. However, the leaching of Cd from MSWIBA washed with TW still exceeded these allowance limits. Based on the results of the EN leaching test, both alkaline washing solutions (SLW and WW) produced comparable final outcomes for removing Cd contained in MSWIBA. The results from the US TCLP leaching test showed that

washing MSWIBA with WW was more effective than washing with SLW when the L/S ratio was higher than 5. On the other hand, the effects of L/S ratio and washing duration on the reduction of Cd were not apparent.

The leaching results of Se are shown in Fig. 5. It can be seen from the EN leaching test results that SLW and WW were not as effective as TW in reducing the leaching of Se from the MSWIBA. Results from the US TCLP leaching test indicated different performance of the washing solutions. According to the US TCLP leaching test, Se measured in the leachates of WW washed MSWIBA was the highest with L/S ratio of 2.5 but became the lowest with higher L/S ratios, i.e., 5 and 10 in washing.

Sb can exist in four oxidation states (− III, 0, III, and V) of which antimonite (Sb(III)) and antimonate (Sb(V)) are the
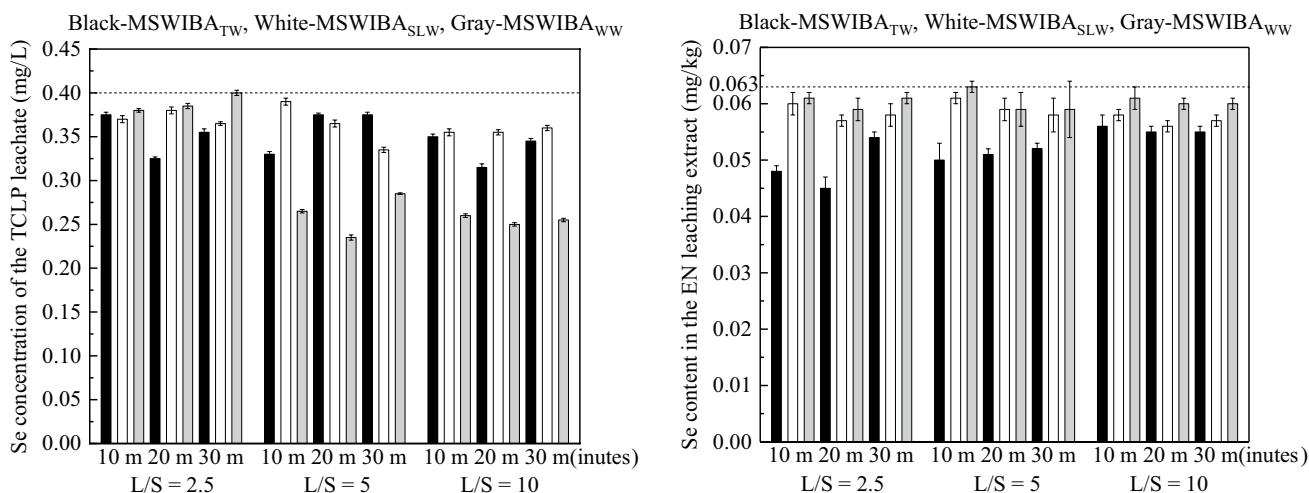
Black-MSWIBA$_{TW}$, White-MSWIBA$_{SLW}$, Gray-MSWIBA$_{WW}$

Black-MSWIBA$_{TW}$, White-MSWIBA$_{SLW}$, Gray-MSWIBA$_{WW}$

Fig. 5 Se leaching concentrations

Black-MSWIBA$_{TW}$, White-MSWIBA$_{SLW}$, Gray-MSWIBA$_{WW}$

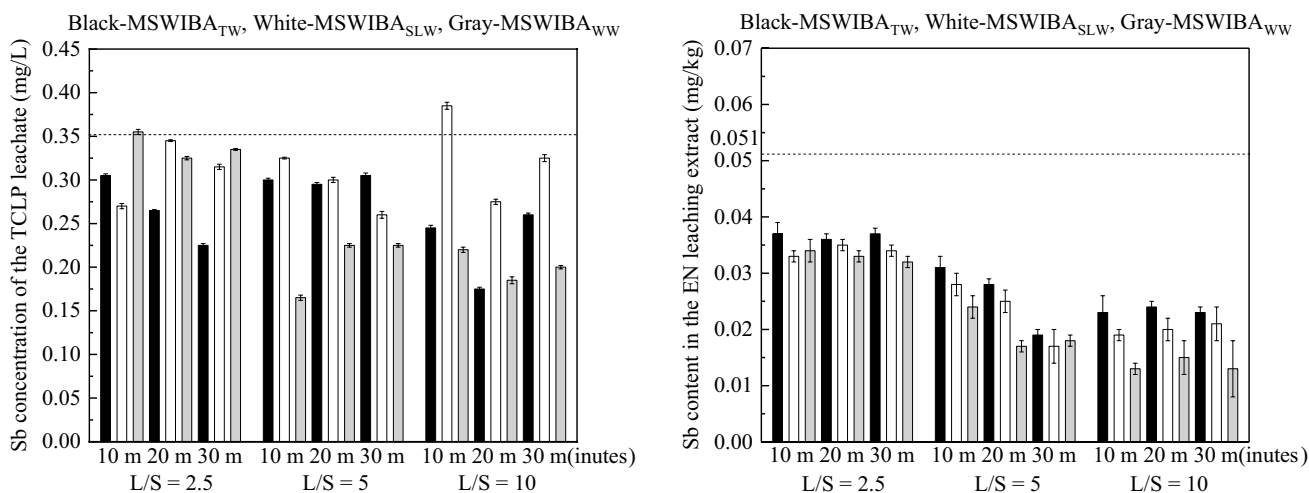Black-MSWIBA$_{TW}$, White-MSWIBA$_{SLW}$, Gray-MSWIBA$_{WW}$

Fig. 6 Sb leaching concentrations

most common species. Sb(V) exists in majority in MSWIBA due to catalyse oxidation of Sb(III) forming Sb(V) during incineration (Paoletti et al. 2001; Zyryanov et al. 2006). According to results shown from the EN leaching test shown in Fig. 6, Sb released from MSWIBA residues after washing with SLW or WW were less compared with washing with TW. WW washing at a higher L/S ratio, i.e., 5 or 10, was most effective in reducing Sb leaching under the US TCLP.

Discussion

Based on the above experimental results, the washing treatment chosen could reduce substantial amounts of heavy metals and anions leaching from the MSWIBA. L/S ratio was

significant in removing chloride. High L/S ratio of 10 could reduce the chloride leaching concentration from 64,805 to 8606 mg/kg after 20 min of washing with WW. Nevertheless, this level still exceeded the maximum permissible chloride leaching concentration for inert waste which is 800 mg/kg. To reduce sulfate leaching, the most effective washing solution was WW. The best performance was achieved with a L/S ratio of 10. Unfortunately, the emission of 1600 mg/kg still exceeded the limit of 1000 mg/kg for inert waste. For the treatment of heavy metals, washing with SLW or WW could reduce the leaching of Cd from the MSWIBA to a level lower than the regulatory limit for inert waste. Therefore, the optimal washing treatment identified in this study is applying WW at a L/S ratio of 10, thereby turning the MSWIBA to a non-hazardous waste, though not an inert waste.

In order to gain insight into the mobility of contaminants, the oxide compositions and total metal contents in samples before and after washing by TW, SLW, and WW, respectively, for 20 min at the optimal L/S ratio of 10 were measured with results shown in Table 5. Compared with the raw MSWIBA, the contents of potassium and sodium in the washed samples were significantly reduced, indicating that most potassium and sodium salts existed in water-soluble forms. To the contrary, calcium and iron concentrations increased after washing with SLW or WW, likely due to some calcium and iron salts depositing in the pores of the MSWIBA during washing. On the other hand, total metal content in the MSWIBA before and after washing did not change too much, indicating retention and possibly stabilization of metals during the washing process.

Figure 7 shows the changes in crystalline phases in the MSWIBA samples at various stages (raw, after washing, and after leaching tests). The crystal phases of the raw MSWIBA mainly included calcite $Ca(CO)_3$ (PDF#86–0174), calcium silicate $CaSi_2O_5$ (PDF#51–0092), and silicon oxide SiO_2 (PDF#88–2488). After washing, the formation of new crystal phases which was conducive to the stabilization of some heavy metals in the ash was detected. As irons were introduced from WW during washing, As-Fe co-precipitation to form pharmacosiderite $4Fe_4(OH)\cdot3(AsO_4)\cdot5(H_2O)$ (PDF#74–1496) occurred, thereby immobilizing As in the ash. Cadmium was also stabilized during washing due to the formation of cadmium hydroxide sulfate $Cd_2(OH)_2SO_4$

(PDF#75–2314). Likewise, Sb species were precipitated in SLW and WW during washing forming insoluble and stable hydromeite $3CaO\cdot2Sb_2O_5\cdot8H_2O$ (PDF#02–1387) under high pH and calcium availability. To the contrary, no crystalline phases of Se were found after washing for inhibiting the leaching of Se. Hanum et al. (2018) observed that the addition of $Ca(OH)_2$ to coal fly ash could suppress the leaching of many elements including As, B, and F, except Se. This result indicated that calcium cations were not effective in stabilizing Se ions. Zhang et al. (2016) reported the leaching concentrations of Se from MSWI fly ash reaching the peak at pH 11. Sun et al. (2017) found that Se sorption decreased as pH increased from 2.5 to 9.5 and then maintained at 40% level at pH between 9.5 and 11.5. Mulugeta et al. (2010) observed high concentrations of Se in the leachates of pH 7.2–11.3, but low concentrations of Se in the leachates with a pH mildly less than 7. As explained above, Se exists mainly as $Se^{IV}O_3^{2-}$ and $Se^{VI}O_4^{2-}$ oxyanions in alkaline conditions and alkalinity could enhance the mobility and movement of Se oxyanions. All the above mineral precipitates remained stable during the TCLP and EN leaching tests, as evident from results of the XRD tests on the solid residues of the leaching tests shown in Fig. 7. Consequently, leaching concentrations of As, Cd, and Sb reduced significantly after washing by SLW and WW.

The leaching problems of As, Cd, Se, Sb, chloride, and sulfate with MSWIBA were significantly reduced by the special washing treatment of the MSWIBA. After treatment in

Table 5 Oxide compositions and total metal contents in MSWIBA before and after 20-min washing at L/S ratio of 10

|	MSWIBA$_{RAW}$	MSWIBA$_{TW}$	MSWIBA$_{SLW}$	MSWIBA$_{WW}$
Na$_2$O	3.14±0.05	0.63±0.15	0.53±0.08	0.64±0.09
MgO	2.51±0.14	3.13±0.10	1.98±0.09	2.43±0.09
Al$_2$O$_3$	10.83±0.15	12.81±0.11	8.13±0.14	9.72±0.13
SiO$_2$	37.65±0.10	38.65±0.08	32.48±0.09	36.10±0.09
Fe$_2$O$_3$	6.63±0.01	6.98±0.05	6.10±0.12	8.05±0.13
P$_2$O$_5$	3.52±0.02	3.98±0.12	3.26±0.05	3.38±0.13
TiO$_2$	1.27±0.02	1.54±0.02	1.68±0.02	1.61±0.02
K$_2$O	1.77±0.020	0.38±0.05	0.41±0.02	0.44±0.013
CaO	24.64±0.10	25.60±0.10	39.30±0.10	34.50±0.10
SO$_3$	4.55±0.05	1.31±0.07	1.89±0.09	1.09±0.07
LOI	2.52±0.01	3.20±0.01	4.10±0.01	1.20±0.01
As	21.82±0.88	20.92±0.79	20.43±1.03	22.38±2.11
Ba	1170.53±58.15	1003.33±47.13	1054.84±52.41	1038.20±260.94
Cd	78.55±1.62	76.30±1.47	73.58±3.70	72.41±2.64
Cr	323.00±15.94	313.26±13.03	311.52±15.38	312.08±115.23
Cu	1638.53±30.44	1608.50±74.19	1595.92±296.46	1612.20±210.38
Ni	111.71±5.49	102.73±3.01	99.80±4.90	95.50±0.11
Pb	682.12±33.96	647.00±32.06	654.38±32.58	620.43±0.27
Sb	3.37±0.16	3.23±0.04	3.30±0.19	3.17±0.41
Se	4.69±0.18	4.43±0.03	4.40±0.30	4.16±0.08
Zn	6701.93±806.71	6649.06±75.05	6601.20±794.59	6683.50±471.71
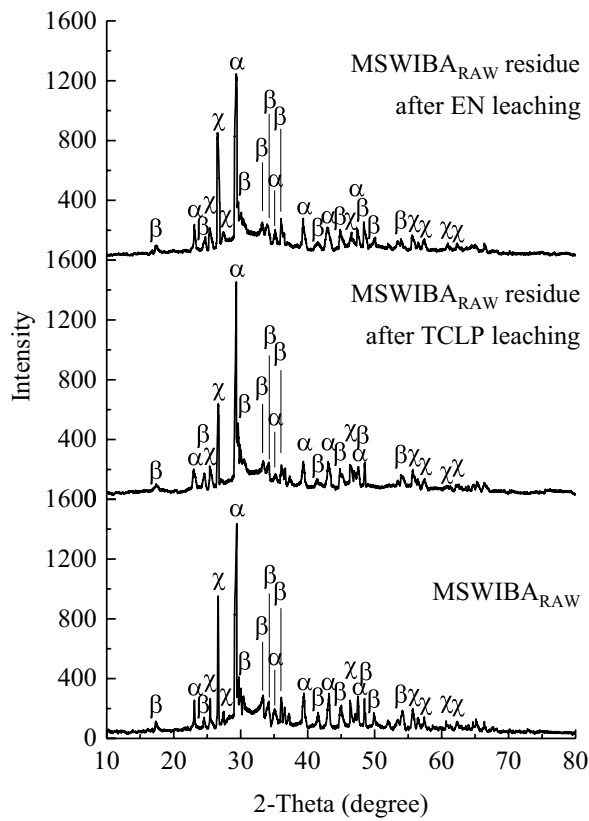
(a)
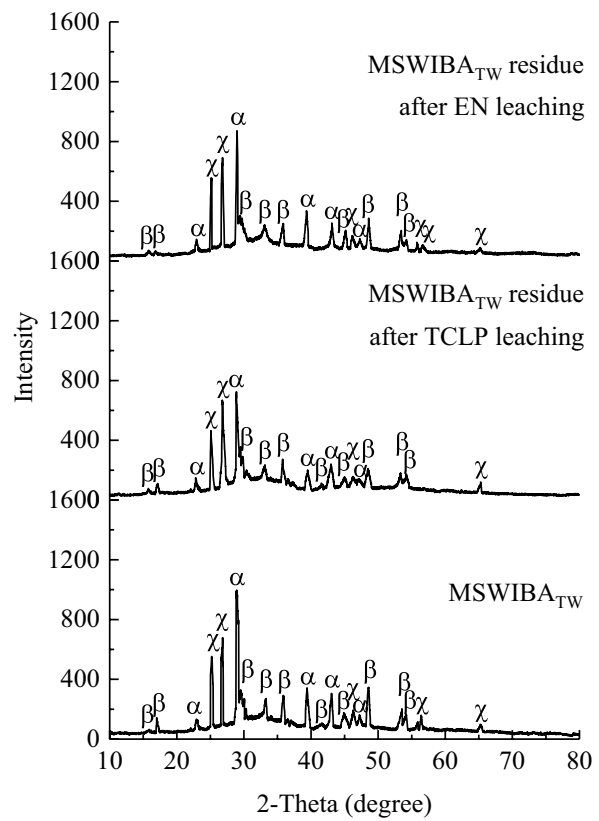
(b)
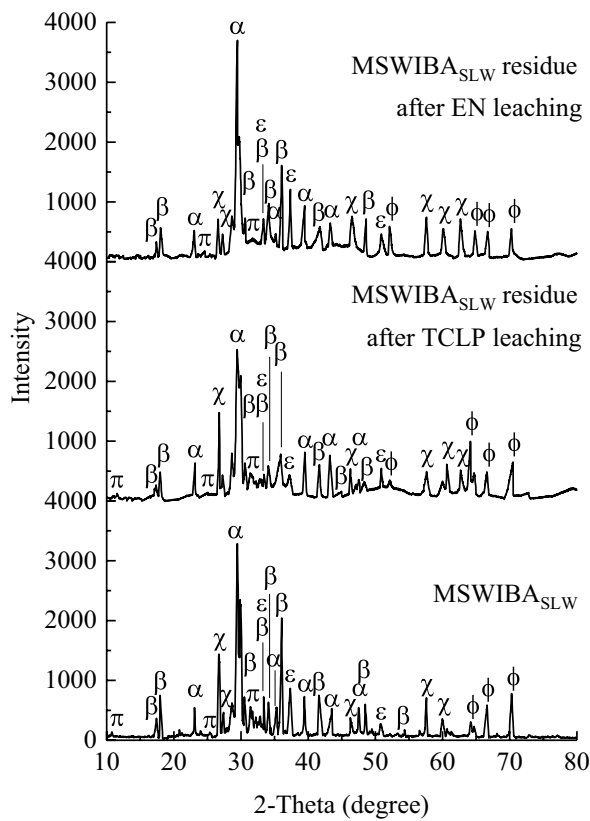
(c)
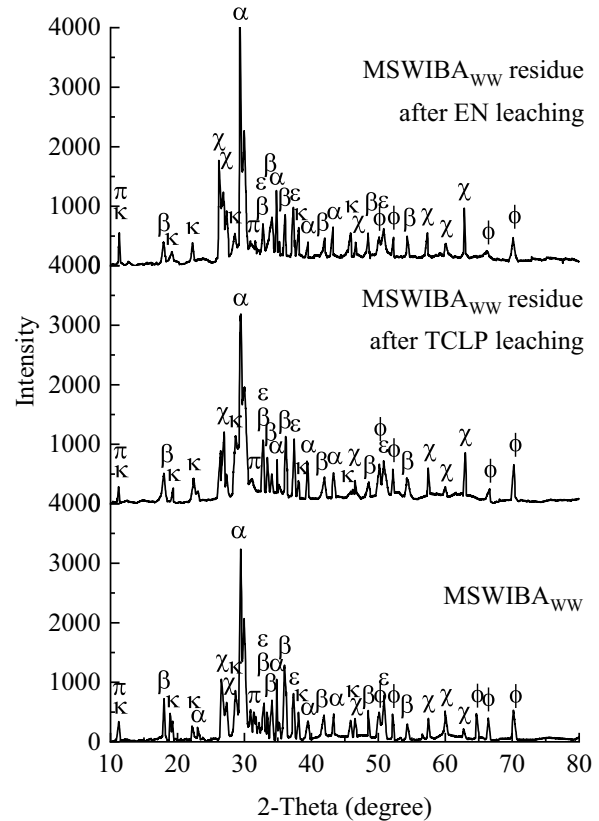
(d)

◄**Fig. 7** XRD crystalline phases in raw, washed, and tested MSWIBA samples. (α, calcite; β, calcium silicate; χ, silicon oxide; ε portlandite; ϕ, hydromeite; π, cadmium hydroxide sulfate; κ pharmacosiderite)

the optimal conditions, i.e., WW washing at the L/S ratio of 10 for at least 20 min, As, Cd, Se, and Sb leaching concentrations fell below the acceptable limits for disposal in inert waste landfills specified in European Council Decision 2003/33/EC (2003). Unfortunately, the chloride and sulfate leaching, though dropped to 8606 and 1600 mg/kg, respectively, still failed to meet the limits of 800 and 1000 mg/kg, respectively, for disposal in inert waste landfills. For this reason, the treated MSWIBA cannot be directly recycled as an engineering material in view of the potential environmental risks. It is therefore desirable to either explore feasible means for improving the washing treatment or to identify other ways of recycling the ash so treated with WW. The management of solid waste together with related environmental protection standards in the Netherlands is widely recognized and adopted for reference by other countries. According to the The Dutch Building Material Decree (1999), the treated MSWIBA should meet the chloride and sulfate leaching requirements (chloride limit of 8800 mg/kg and sulfate limit of 20,000 mg/kg) for use in infrastructure projects such as ICM (isolation, control, and monitoring) building materials.

Conclusion

This study has demonstrated the use of three different washing solutions for the treatment of MSWIBA to reduce its harmful leaching of constituents including chloride, sulfate, As, Cd, Se, and Sb to the environment. The raw MSWIBA used in this study was classified as a hazardous material due to its high chloride concentration. Experiments conducted indicated that simple washing with tap water or alkaline water could largely remove chloride from the MSWIBA. This effect was more notable with high L/S ratios. At a L/S ratio of 10, the chloride leaching concentration was reduced to below 15,000 mg/kg, turning the ash to non-hazardous waste, and yet, it was still not accepted as an inert material. WW was the most effective solution for reducing the leaching of sulfate. It was noted that WW washing for 20 min at L/S ratio of 10 could remove about 83% of sulfate from the MSWIBA. According to the results from the US TCLP leaching test, WW is a promising washing solution which could reduce the leaching of As, Cd, Se, and Sb from the MSWIBA into acidic environment. However, this effect of WW was only notable when L/S ratio was higher than 5. After treatment, the subsequent leaching of heavy metals could be reduced to below the limits for inert waste. It is desirable for washing solutions to contain calcium and iron,

which could contribute to stabilization of some metal species, preventing them from leaching out from MSWIBA. Wastewater from concrete plants is a preferred washing solution for reducing the leaching of heavy metals from MSWIBA given its high efficiency. According to the European Council Decision criteria, chloride and sulfate leaching from the washed MSWIBA exceeds the acceptance levels for inert waste. However, the treated ash may be suitable for use as ICM building materials under the criteria stipulated in the Dutch Building Materials Decree.

Declarations

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