

Review Article

Review on Cement Stabilization/Solidification of Municipal Solid Waste Incineration Fly Ash

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Municipal solid waste incineration (MSWI) fly ash must be treated properly prior to being disposed in the security landfill due to its serious pollution toxicity. Nowadays, lots of studies have demonstrated that cement-based stabilization/solidification could reduce the toxicity pollution effectively by encapsulating the heavy metals into cement matrix, which leads to greater capacity and weight. This paper compares and discusses the MSWI fly ash treatment with the mostly used matrix materials such as Portland cement, phosphate cement, aluminate cement, and alkaline activated cement. Moreover, immobilization mechanism introduced by the interaction between the MSWI fly ash and hydrated cement matrix materials, such as the physical cementing effect, adsorption, isomorphous replacement, and complex precipitation, was explored in depth. The paper also pointed out some reasonable development directions for cement-based stabilization/solidification technology to improve the effectiveness and application of cement-based stabilization/solidification technology.

1. Introduction

Waste incineration technology has been widely used globally because it can achieve the goal of harmlessness, waste minimization, and resource utilization of municipal solid waste incineration. Nonetheless, there will be a large amount of MSWI fly ash coupled with various types of toxic heavy metal materials with high concentration after the incineration, accounting about 3 to 5 percent of the total waste, which not only has severely restricted the enterprise's survival and development, but also causes a great threat to the human survival environment and health. Therefore, disposing MSWI fly ash and solving the problem of heavy metal pollution properly is a task which brooks no delay.

Nowadays, cement-based stabilization/solidification technology has been widely used all over the world for 60 years, it and has become an economic efficiency approach to immobilize toxic and harmful heavy metal in the MSWI fly ash by being fixed in the hydration products, such as calcium silicate hydrate gel (C-S-H) and ettringite (AFt) which were formed by chemical reaction between cement and water [1–4]. Besides, the advantages of cement-based stabilization/solidification not only have simple operation and low processing cost, but also significant reduction of percolation of heavy metal ions in the hazardous waste landfill, which could meet the Standard for Pollution Control on the Security Landfill. Thus, the United States Environmental Protection Agency (EPA) has considered the method as the best way to deal with poisonous and harmful waste in the world so far [5].

On the basis of existing research, this paper made a comparative analysis on the effect of MSWI fly ash immobilized with different types of cementitious materials, such as Portland cement, phosphate cement, aluminate cement, and alkaline activated cement, and then further explored the influence factors and immobilization mechanism through the interactions between cement hydration products and heavy metals. Moreover, some suggestions were also put forward on the application and development of cement-based stabilization/solidification technology in the next studies.

2. Characteristics of MSWI Fly Ash

Comparative analysis of MSWI fly ash around the world showed that the composition of MSWI fly ash was relatively complicated. It not only consisted of silicate, chlorine salt, calcium hydroxide, and sulfate, but also absorbed a lot of the poisonous and harmful substances in the flue gas, including dioxins, furans, mercury (Hg), copper (Cu), lead (Pb), manganese (Mn), cadmium (Cd), chromium (Cr), nickel (Ni), and so on [6, 7], which had already become a serious threat to the ecological environment, food safety, and human health. However, the organic matters would decompose and evaporate during the incineration process, and its content in the MSWI fly ash was generally below 2%, thus it meant that the treatment and remediation of heavy metals had become the main target in the stabilization/solidification process of MSWI fly ash. Moreover, once heavy metals caused harm to human health and natural environment, it would be serious, long-term, latent, cumulated, and irreversible [8-10]. Therefore, finding the solutions to solve the heavy metal pollution of MSWI fly ash has become a top priority. Tables 1 and 2 summarize the sources and chemical speciation of heavy metals in the MSWI fly ash, which could provide some theory guidance for the MSWI fly ash immobilization study in the following.

3. Effect of Different Cementitious Matrix Materials on MSWI Fly Ash Immobilization

3.1. Portland Cement. Lombardi et al. and Polettini et al. [11, 12] reported that cement could effectively immobilize contaminants, but the compressive strength will be reduced and leaching concentration will be increased when the MSWI fly ash addition is increased. Also, Dermatas and Meng [13] pointed out that the most obvious shortcoming of cement stabilization/solidification was high capacity, weight increased rate, and poor long-term stability. The capacity and weight increased rate was up to 2.0. However, when the MSWI fly ash was prewashed with water, phosphoric acid, or other solutions before cement-based stabilization/solidification, it would drastically reduce the toxicity of heavy metals and reduce the consumption of cement. However, there was still tricalcium aluminate (C_3A) in the cement solidified body after the 28-day curing period, but C₃A would greatly reduce its compressive strength [14], and it was difficult for Portland cement solidified body to achieve high mechanical strength whether the MSWI fly ash was pretreated or not [15].

What is more, some literature studies proposed that some heavy metals in the MSWI fly ash such as Cr⁶⁺, Cd²⁺, Pb²⁺, and Cu²⁺ could dramatically accelerate C₃S hydration whether it was in the late or initial stage of hydration reaction, but the Zn²⁺ only promoted the C₃S hydration during the later period of hydration [16, 17]. But some studies reported that heavy metals would delay the cement hydration to some extent. In detail, Cu²⁺ and Pb²⁺ usually bound on the surface of cement clinker particles in the form of insoluble salt, such as carbonates and sulfates, which inhibited the cement hydration. Ma et al. [18] proposed that Cu, Zn, and Pb could not obviously change the cement hydration process according to the analysis of XRD spectrum, and Cu, Zn, and Pb were immobilized within cement hydration products through a series of effect mechanism, such as physical fixation and adsorption mechanisms. Vogg et al. [19] pointed out that heavy metals would leak out little by little in an acidic environment, but there would be no

detection of Cd, Cu, Pb, and Zn under the condition at pH value of 10.0.

3.2. Phosphate Cement. Magnesium phosphate cement was also used to immobilize MSWI fly ash, the reason of which was not only the alkalescence of potassium phosphate cement with the characteristic of dense structure and high strength, but also the widespread use of phosphate as curing agents. It was found that the compressive strength and leaching toxicity for magnesium phosphate cement on Cr³⁺, Pb²⁺, Ni²⁺, Cd²⁺, and Zn²⁺ could meet the national corresponding standards [20-22]. On the one hand, there were some macropores and gassy surface in the light burned magnesia powder crystal with a high activity, which brought the strong adsorption for heavy metals. On the other hand, magnesia powder with higher activity could give impetus to the violent reaction with magnesium salt solution and chemical precipitation of heavy metals [23]. Therefore, it was easy for magnesium hydroxide to absorb and remove heavy metals from industrial waste water under the help of its high specific surface area and adsorption capacity.

Besides, the solidification effect on Pb²⁺ of magnesium phosphate cement would be more prominent in contrast with other heavy metals. It was the results from both chemical reaction and physical encapsulation. On the one hand, Pb^{2+} was more likely to react with PO_4^{3-} and HPO₄²⁻, and then generate insoluble phosphate precipitation containing Pb. Moreover, the heavy metals could be encapsulated securely because of compact reticular structure of magnesium phosphate cement, in which the radioactive elements can be solidified effectively as well [24-26]. And the heavy metals such as Pb, Cd, and Cr almost could not be detected when the addition of magnesium potassium phosphate cement was up to 40%. Besides, it was noted that the leaching concentration of heavy metals always followed a descending order of Cu > Zn > Ni > Pb > Cd > Cr when eluviated in different leaching environments.

3.3. Aluminate Cement. There were very few studies on the heavy metals immobilization in aluminate cement and sulphoaluminate cement. But Sahu et al. and Luo et al. [27-29] found that sulphoaluminate cement could also reduce the leaching toxicity of heavy metals, and it might be prone to fix Pb²⁺ than Zn²⁺, Cd²⁺, and Cu²⁺ due to the selectivity of its hydration products. Besides, the heavy metals mainly deposited in the complex chemical speciation. For example, Pb²⁺ is mainly in the form of 2PbCO₃·Pb(OH)₂; Cd²⁺ mainly exists in the form of CdPbO₃. Overall, the adsorption stability of Pb²⁺, Cu²⁺, Cd²⁺, and Zn²⁺ immobilized by sulphoaluminate cement is good, especially for Pb²⁺.

3.4. Alkaline Activated Cement. Alkaline activated cement is a kind of cementious material formed by reaction of the alkaline activator and materials with volcanic activity or latent hydraulicity. The chemical reactions including the precipitation of calcium silicoaluminate gel (CASH) and sodium

Heavy metal	Source		
Pb	Gasoline additives, ammunition, solder, paint, pesticide		
Hg	Electrical manufacturing, pharmaceuticals, paints, plastics, paper, batteries, coal, mercury smelters, and mercury		
	preparation plants		
Cd	Plastic, enamels with pigment, electroplating, metal covering		
Cr	Mine, smelting plant, electroplating factory, chrome tanning system		
As	Atmospheric dust, tailings and pesticide, leather, chemical pharmaceutical, metallurgy, decolorant		
Cu	Mine, metal processing, machinery manufacturing, steel production		
Zn	Galvanizing, smelting, machinery manufacturing, organic synthesis, mine		

TABLE 2: Chemical speciation of heavy metals in MSWI fly ash.

Heavy metal	Speciation
Pb	PbCl ₂ , PbCO ₃ , PbO, Pb ₃ SiO ₃ , Pb ₃ SiO ₄ ,Pb ₃ O ₂ SO ₄ ,
10	$Pb_3Sb_2O_7$
Cd	Cd(OH) ₂ , CdO, CdSO ₄ , CdCl ₂ , CdSiO ₄
Zn	ZnCl ₂ , ZnO, Zn(OH) ₂ , ZnCO ₃ , ZnSO ₄ ,
211	K_2ZnCl_4
Fe	Fe ₂ O ₃ , Fe ₃ O ₄
Cu	Cu(OH) ₂ , CuO, CuCO ₃

silicoaluminate gel (NASH) may occur during the process of setting and hardening, then forming a three-dimensional aluminum silicate network structure which consists of AlO_4 and SiO_4 tetrahedral structure units, which could be used to seal the contaminants tightly in the cavity [30, 31].

Zak and Deja [32] used chemical reagents to synthesize C-S-H, CASH, and NASH, and then found that Zn^{2+} , Cd^{2+} , Pb^{2+} , Cr^{3+} , and Cr^{6+} inhibited the synthesis of C-S-H, CASH, and NASH structures, and C-S-H and NASH could immobilize heavy metal at the degree of 99.9% and 93.1%, respectively. It could be deduced that the alkaline activated cement could be used to seal heavy metals, which was in line with the conclusion from Davidovits [33]; meanwhile, he also stated that the network skeleton of alkaline activated cement was of perfect stability even though under nuclear radiation.

From the above, cement-based stabilization/solidification could effectively fix the heavy metal in the cement with the advantages of simple operation, low cost, and high strength of cement solidified body, but it also gave rise to poor stability, high capacity, and weight increased rate. Furthermore, it should be noted that the cement-based stabilization/ solidification of MSWI fly ash just belonged to temporary concentration control, which could not guarantee its longterm security and stability, especially under the acid environment. Moreover, it was indeed feasible to fix the MSWI fly ash with cement when mixed with little MSWI fly ash, but it was hard to ensure that the performances of cement solidified body meet the national landfill standards with increased MSWI fly ash. If there were special salts in the MSWI fly ash, the salts would hinder the cement-hardening process and weaken its mechanical strength, leading to high leaching rates of heavy metals.

Comparative analysis of the effect of different cementitious matrix materials on MSWI fly ash immobilization was summarized in Table 3. There are lots of smaller capillary pores in the magnesium phosphate cement,

TABLE 3: MSWI fly ash immobilized with different cementitious matrix materials.

Cement	Advantages	Disadvantages
	(1) Simple operation	(1) High capacity and weight increased rates
Portland cement	(2) Low cost	(2) Poor long-term security and stability
	(3) Moderate compressive strength	
Phosphate	(1) High compressive strength	(1) High capacity and weight increased rates
cement	(2) Good dry shrinkage(3) Low cost	(2) Poor long-term security and stability
Aluminate	(1) Simple operation	(1) High capacity and weight increased rates
cement	(2) Low cost	(2) Poor stability(3) Poor dry shrinkage
Alkali activated cement	 (1) High compressive strength (2) Low cost (3) Good long-term security and stability 	 (1) High capacity and weight increased rates (3) Poor dry shrinkage

resulting in a more compact microstructure, thus it could immobilize the heavy metals more effectively in theory. However, what calls for special attention is that magnesium phosphate cement would lose the strength and burst into fragments when exposed to the humid environment for a long time due to the poor water resistance, which would put the disposal of MSWI fly ash at risk. Alkaline activated cement was of the ideal performances, including excellent mechanical properties and durability, whereas shrinkage cracking and weak anticarbonization were the two fatal flaws of this material, which would result in secondary pollution to the environment, ultimately affecting the health of human beings. It can be concluded from these literature studies that the solidification effect of these cementitious materials on MSWI fly ash is alkali activated cement > magnesium phosphate cement > Portland cement > aluminate cement.

4. Immobilization Mechanisms

During the process of cement hydration, heavy metals eventually settled in the cement hydration products in the form of hydroxide or complex by reacting with cement by means of the physical cementing effect, chemical absorption, isomorphous replacement, and complex precipitation. Meanwhile, cement also provides the alkaline environment for solidified body so as to effectively inhibit the infiltration of heavy metals. Besides, these hydration products including C-S-H, AFt, single sulfur hydrated calcium thioaluminate (AFm), and CASH, rather than calcium hydroxide, were difficult to dissolve, and these insoluble hydrated products would play important roles during heavy metal immobilization. Thus, the immobilization of MSWI fly ash in cementbased stabilization/solidification was investigated in depth mainly from these aspects such as hydration products and immobilization effect.

$$C_{3}S + H_{2}O \longrightarrow CaO \cdot SiO_{2} \cdot YH_{2}O(C - S - H) + Ca(OH)_{2}(CH),$$
(1)

$$C_2S + H_2O \longrightarrow CaO \cdot SiO_2 \cdot YH_2O(C - S - H) + Ca(OH)_2(CH),$$
(2)

$$C_{3}A + 6H_{2}O \longrightarrow 3CaO \cdot Al_{2}O_{3} \cdot 6H_{2}O (CAH),$$
(3)

$$C_{3}A + 3CaSO_{4} \cdot 2H_{2}O + 26H_{2}O \longrightarrow 3CaO \cdot Al_{2}O_{3} \cdot 3CaSO_{4} \cdot 32H_{2}O (AFt),$$

$$(4.1)$$

 $3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{CaSO}_4 \cdot 32\text{H}_2\text{O} + 2\text{C}_3\text{A} + 4\text{H}_2\text{O} \longrightarrow 3[3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{CaSO}_4 \cdot 12\text{H}_2\text{O}] \text{ (AFm)}, \tag{4.2}$

$$C_4AF + 7H_2O \longrightarrow 3CaO \cdot Al_2O_3 \cdot 6H_2O(CAH) + CaO \cdot Fe_2O_3 \cdot H_2O(CFH).$$
(5)

4.1. C-S-H. C-S-H is a type of amorphous microporous phase with high specific surface area and high-density hydrogen bonding, which could tightly bind heavy metals with strong chemical adsorption. In general, the C-S-H colloid of rich calcium with positive charges on the surface has preferential adsorption of anions including OH⁻, Cl⁻, SO₄²⁻, and so on, but the C-S-H colloids of rich silicon are preferentially adsorbed cations [34]. Furthermore, it was confirmed that the adsorption of heavy metals is always significantly influenced by pH value, and it would decrease obviously when the pH value surpasses 9.0 [35].

In addition to this, C-S-H had significant cation exchange capacity, and some heavy metals such as Cr^{6+} , Cd^{2+} , Zn^{2+} , and Pb^{2+} often had stronger adsorption affinity in contrast with alkali metal ions. The heavy metals were thus prone to ions exchange, and they would then stabilize in the lattice. Ca^{2+} , Al^{3+} , and Si^{4+} in the cement solidified body are easily replaced by heavy metal cation, and SO_4^{2-} and OH^- can be easily replaced by heavy metal anions [36]. Similarly, Zn^{2+} could replace the Ca^{2+} in C-S-H or react with Ca to produce the oxides or hydroxides containing Ca and Zn [37]. Moreover, Cr^{3+} could be combined into C-S-H by replacing Si^{2+} in C-S-H [38], and CrO_4^{2-} could be fixed in the cement solidified body by replacing SO_4^{2-} .

However, Pb²⁺ in C-S-H cannot replace Ca²⁺, but it can be fixed in C-S-H in the form of hydroxide precipitation and carbonate precipitation, while Pb-C-S-H was observed by means of X-ray photoelectron spectroscopy (XPS) and SEM/EDS in the research of Cocke [39]. It should be noted that weak alkalescent environment could further retard leaching toxicity of heavy metals in cement solidified body in most cases. It should be noted that hydroxide precipitation occurs when the pH value of leaching solution dissolved heavy metals is raised to some optimum level for a specific metal. Table 4 exhibits that the optimum pH value is always different for each metal and for different valence states of a single metal. For example, the leaching rates for Cd^{2+} decreased when the pH value is more than 8.0, and Pb^{2+} leached to the least when the pH value is 10.3 [40, 41].

4.2. AFt and AFm. AFt phase, $3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$, is one of the main hydration products of cement, but it can be easily changed into AFm (Ca₂Al(OH)₆·0.5X·H₂O) when chloride or sulfate is insufficient. As shown in Figure 1, AFt phases are chemically and mineralogically complex, in which columns of Ca₆Al(OH)₆·24H₂O are lines of Al(OH)₆³⁻ octahedrally bonded with three calcium polyhedrons, and each calcium polyhedron is with OH⁻ and four water molecules, which presents an orientational column structure on the whole.

In most cases, AFt can absorb and immobilize the heavy metals through the isomorphous replacement and chemical adsorption; a plenty of studies demonstrated that AFt have a very strong lattice binding effect on heavy metal ions. For example, Cs, Sn, and other metal ions could also be adsorbed on the cylinder surface of AFt [42, 43]. On the one hand, Cd²⁺, Ni²⁺, Zn²⁺, Pb²⁺, and Co²⁺ could replace Ca²⁺, and Cr³⁺, Ti³⁺, Mn^{3+} , Si⁴⁺, and Fe³⁺ can replace Al³⁺ in AFt; on the other hand, Cl⁻, CO₃²⁻, SeO₄²⁻, BrO₃⁻, AsO₄²⁻, and ClO₃⁻ can replace SO_4^{2-} [44–47]. Besides, Sabine and Lan et al. found that Cr^{3+} , CrO_4^{2-} , and $Cr_2O_7^{2-}$ can get inside the AFt lattice and then stabilize in the AFt phases. Tashiro et al. [48-50]found that heavy metal oxides and hydroxides could promote the formation and crystal growth of ettringite and produce some changes in microstructure and Cr2O3 and Cu(OH)2 exert a considerable influence. Similar to AFt, Ca²⁺, Al³⁺, CO₃²⁻, and SO₄³⁻ in AFm phases are easy to be replaced by other ions, and AFm have a more stable structure than AFt. Several studies have been put forward to explain that AFm could effectively reduce the content of heavy metal ions in the water, such as Cr³⁺, CrO₄²⁻, Cd²⁺, B³⁺, Se⁶⁺, and so on [44, 51, 52]. Advances in Materials Science and Engineering

W

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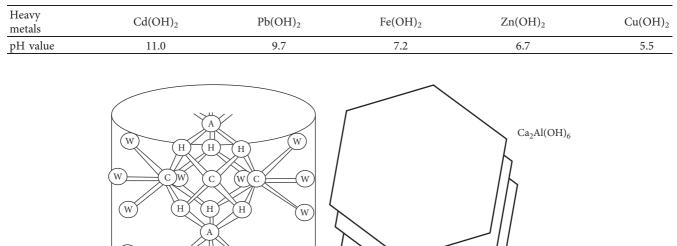
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Н

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TABLE 4: Optimum pH value of heavy metal precipitates.



W

w

FIGURE 1: Cellular structure of AFt and AFm.

Isomorphous replacement is also known as isomorphous substitution. In mineral crystallization, the position occupied by some ions or atoms in the crystal structure is partly occupied by other ions or atoms of similar nature and size, but the crystal structure is basically unchanged. The electronegativity of Cr^{3+} and Al^{3+} is close, and both ions' electricity price is the same, so it can replace the calcium alum stone Al^{3+} form relatively stable substitutional solid solution. On the contrary, Cr^{6+} would change the molecular symmetry and cause a great influence on the lattice structure of AFt when entered into the lattice structure. Therefore, AFt is prone to solidly immobilizing Cr^{3+} than Cr^{6+} .

4.3. CASH. CASH is a kind of zeolite minerals composed of oxygen-silicon tetrahedron, which has strong ability of adsorption and ion exchange capacity due to its higher specific surface area with $500 \sim 1000 \text{ m}^2/\text{g}$. However, Si⁴⁺ in the oxygen-silicon tetrahedron always be substituted by Al³⁺ and form a alumina tetrahedron with some extra negative charge, which needs alkali metal cations to achieve the charge balance. There is poor binding force between alkali metal cations and the CASH crystal, because most of the alkali metal cations are located in a cavity or pore structures. Besides, these heavy metal ions including Cr³⁺, Cd²⁺, and Pb²⁺ have stronger adsorption ability than alkali metal cations in CASH, so the heavy metal ions are more likely to be effectively immobilized in CASH.

5. Summary and Prospect

(b)

AFm

The comparative analysis of the solidification effect and leaching toxicity of MSWI fly ash treated by various types of cement has been investigated and discussed in depth in this paper and found that it was more effective to use the phosphate cement and alkali activated cement to immobilize the heavy metals. And this paper also looks at the interaction of cement hydration products and heavy metals in particular. However, there are also lots of controversies about immobilization mechanism of heavy metals with cementbased stabilization/solidification technology. For example, these immobilization mechanisms always change with external environment and matrix materials structure, and meanwhile, the heavy metal hydrolyses always give rise to variation of pH values and cement hydration process, which would make the study on immobilization mechanisms and hydration products fixing effects of heavy metal more complex. But beyond all that, there are few in-depth studies on the long-term safety and stability after the landfill of MSWI fly ash solidification and no universal practical method to realize the reutilization of MSWI fly ash. Therefore, it is to be hoped that the proper treatment and disposal of MSWI fly ash would be realized with the efforts in the next few years.

Conflicts of Interest

The authors declare no conflicts of interest.

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