Waste Disposal & Sustainable Energy (2022) 4:69–74
https://doi.org/10.1007/s42768-022-00102-6

VIEWPOINT

Low-carbon stabilization/solidification of municipal solid waste incineration fly ash

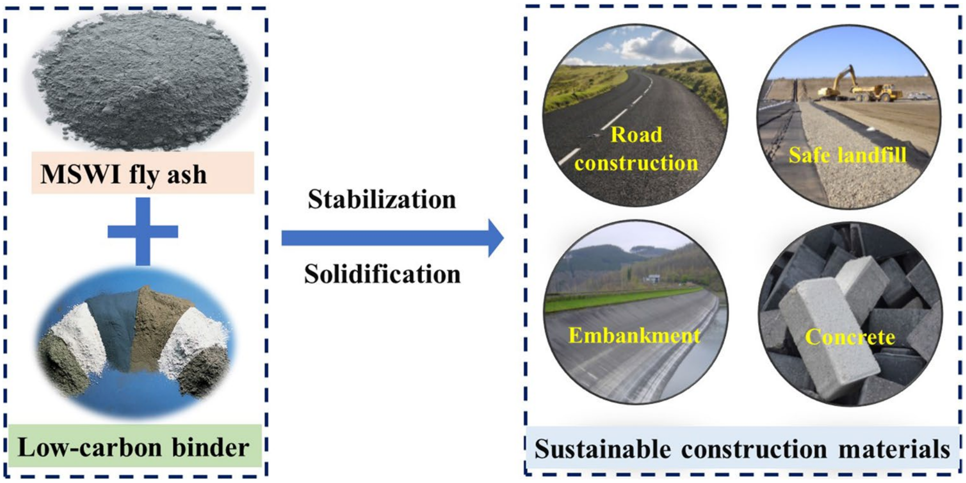
Chen Sun[1] · **Lei Wang**[1] · **Xiaoqing Lin**[1] · **Shengyong Lu**[1] · **Qunxing Huang**[1] · **Jianhua Yan**[1]

Received: 12 May 2022 / Revised: 12 May 2022 / Accepted: 16 May 2022 / Published online: 2 June 2022

Abstract

Municipal solid waste incineration (MSWI) fly ash (FA) is classified as hazardous waste, which requires additional treatment before disposal or further utilization. Stabilization/solidification (S/S) is regarded as a low-cost and high-efficient method for MSWI FA treatment. “Low-carbon S/S” has captured extensive interest in recent years, which could treat hazardous wastes and enable resource recycling in a sustainable way. This article introduced the state-of-art low-carbon S/S strategies for MSWI FA treatment. The immobilization mechanisms of pollutants in various matrices were also discussed. Prospects were raised to foster the actualization of sustainable management of MSWI FA.

Graphical abstract



Keywords Municipal solid waste incineration fly ash · Stabilization · Solidification · Potential toxic elements · Hazardous waste treatment · Green and sustainable remediation · Carbon neutrality

Introduction

Rapid urbanization and fast economic development result in a significant increase in municipal solid waste. According to the prediction [1], solid waste from cities alone will increase to 2.2 billion tons in 2025. Incineration has been the mainstream approach for MSW (municipal solid waste) treatment. The amount of MSW treated by incineration in China increased from 11.4 million tons per year to 101.8 million tons per year from 2006 to 2018 [2]. The incineration residues usually contain bottom ash (BA) and fly ash (FA). The FA generation equals about 3%–15% of the MSW incinerated, which means that approximately 3.7–18.3 million tons of MSWI (municipal solid waste incineration) FA would be generated per year in China. The

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✉ Lei Wang
wanglei.leo@zju.edu.cn

[1] State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China

FA amount is highly correlated with the furnace type. The fluidized bed furnace produces more FA compared with the moving gate furnace [3]. Due to high contents of potential toxic elements (PTE) and organic pollutants, MSWI FA is classified as hazardous solid waste globally [4]. Various PTEs, such as Zn, Pb, Cu, As, Cr, Cd, and Ni in MSWI FA are at high levels both in total content and leaching solution. In addition, furans, dioxins, and polycyclic aromatic hydrocarbons could be detected in MSWI FA [5]. PTEs immobilization and dioxin degradation are two major challenges that need to be solved before landfill or resource utilization.

The major chemical components of MSWI FA are CaO, SiO_2, Al_2O_3, Na_2O, K_2O, and Fe_2O_3 and chlorine compounds [6]. A highly alkaline environment (pH 10.5–13.5) was generally observed in MSWI FA due to the spray of lime or sodium carbonate for flue gas purification [7]. Reactive aluminosilicate in MSWI FA endows it with potential pozzolanic properties. MSWI FA with micro-scale fine particles (<300 μm) offers a large reactive surface area for the reaction [8]. These characteristics of MSWI FA imply that MSWI FA could be recycled as construction materials.

Stabilization/Solidification (S/S) is widely used for hazardous waste treatment [9, 10]. After S/S treatment, pollutants (e.g., PTEs) in hazardous wastes are converted to a chemically stable form or solidified in matrices [11], while the S/S products can be recycled into eco-friendly construction buildings [12]. However, ordinary Portland cement (OPC) shows unsatisfactory compatibility with the concentrated pollutants in MSWI FA [12]. In addition, OPC production is associated with large CO_2 emissions [13]. Therefore, low-carbon cementitious materials should be designed for the S/S of MSWI FA.

In this paper, the S/S performance of various low-carbon cementitious materials was introduced in detail. The immobilization mechanisms of PTEs in S/S matrices were discussed. And perspectives on low-carbon S/S technologies for MSWI FA were proposed.

Factors affecting leaching behavior of MSWI FA

The PTEs leaching from MSWI FA to the surroundings may cause serious environmental impacts. The chemical speciations of the major PTEs in the MSWI fly ash are in their oxide, silicate, sulfate, and chloride forms [14]. In MSWI FA, the total concentrations of Zn (320–19,000 mg/kg) and Pb (108–16,800 mg/kg) are relatively high, with high leaching potential (up to 50 mg/L and 25 mg/L for Zn and Pb, respectively) [6]. For other elements, such as Ni, although the total concentration in MSWI FA is high (up to 2000 mg/kg), its leachability is quite low (<1 mg/L). Therefore, S/S binders should be designed for the immobilization of Zn and Pb in MSWI FA. The leaching behaviors of PTEs vary with different ash properties. The dissolved organic matter (DOM) content and sulfate and chloride contents could significantly influence the leaching behaviors of PTEs. For example, the DOM (e.g., fulvic acid) in MSWI FA would increase the Cu leachability by forming a Cu-DOM complex [15]. Sulfate and chloride have adverse effects on PTEs leachability. The sulfate phase is beneficial to stabilizing most PTEs [16]; however, the chloride not only enhances the volatility of PTEs during solid waste incineration but also facilitates the PTEs leaching in the form of the chloride phase [17]. In addition, the particle size of MSWI FA also has a great influence on the leaching behavior. Toxic pollutants tend to concentrate in smaller particles [18, 19], and the surface areas of smaller particles are relatively large, resulting in fast leaching kinetics [20]. In conclusion, to explore the PTEs leaching mechanism of various MSWI fly ash, detailed characterization should be conducted to design appropriate S/S binders.

Cementitious materials for S/S of MSWI FA

Calcium-based cement

Various cementitious binders have been designed for the S/S of MSWI FA. Calcium-based cement, especially OPC, is an extensively used S/S binder. The OPC hydration products consist of tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A), and tetracalcium aluminoferrite (C_4AF) [21]. Calcium silicate hydrate (C–S–H) gel and ettringite (AFt) are crucial for the immobilization of pollutants and strength development [9]. The high contents of chloride and sulfate in MSWI FA have negative effects on the hydration reaction and durability [22]. However, the effects of PTEs on the hydration and properties of OPC were distinctive. Some studies have shown that Cr, Cd, and Cu could accelerate C_3S hydration, whereas Pb and Zn could inhibit the reaction of C_3S [9]. The OPC system has a highly alkaline condition (pH>13), which can alleviate the PTEs leachability from MSWI FA via hydroxide precipitation. However, some amphoteric metals (e.g., Zn) have high leachability under such high alkaline conditions [23]. Therefore, OPC is not suitable for the S/S of some amphoteric metals [24]. OPC system also shows unsatisfying long-term durability. It is reported that the leaching potentials of PTEs in OPC-treated MSWI FA increased a lot after 6 years of natural aging [25]. In addition, OPC obtained from the calcination of limestone and clay at 1400 °C leads to abundant carbon emissions.

Calcium aluminate cement (CAC) is a novel calcium-based cement for S/S of MSWI FA [26]. The hydration products of CAC are hexagonal calcium aluminate hydrates (CAH_{10} and C_2AH_8) and cubic hydrogarnet (C_3AH_6). CAC has been used as an efficient alternative binder for stabilizing Pb-contaminated soil, in which Pb partially replacement in the crystalline hydration products could be the major immobilization mechanism [26]. It has been reported that CAC exhibited high compatibility with MSWI FA and high compressive strength due to the fast reaction process and the formation of monocarboaluminate [27]. Compared to the pH (>13) in the OPC system, CAC has a moderate pH (10.5–11) [28], which favors the stabilization of amphoteric metals. Phosphate, as an effective additive, could be incorporated into the CAC system to improve the S/S performance [13, 29]. The phosphate-enhanced CAC system exhibited excellent immobilization efficiency (98.5%) of Pb compared with the OPC system (89.4%). PTEs were immobilized via the chemical formation of insoluble phosphate precipitates and physical encapsulation within the high-density crystal lattice [30].

Magnesium-based cement

Magnesium-based cement is a low-carbon binder family, due to the low calcination temperature of magnesite (650–800 °C) [31]. Compared to OPC calcination at 1400 °C, Magnesium-based cement calcination generated less CO_2 emission [32]. Besides, MgO-derived blocks could sequester abundant CO_2 in their service life [33]. The internal pH of magnesium-based cement ranged from 9 to 12, which is more appropriate for amphoteric toxic metals compared to OPC [34].

Magnesium phosphate cement (MPC) is a fast-hardening magnesium-based cement. MPC is one of the most effective binders for the S/S of MSWI FA due to its strong chemical stability and high bonding strength. The major hydration products of MPC could primarily be divided into $MgNH_4PO_4 \cdot 6H_2O$ [35] and $MgKPO_4 \cdot 6H_2O$ [36], respectively. The solubility product constants (Ksp) of the $MgNH_4PO_4 \cdot 6H_2O$ and $MgKPO_4 \cdot 6H_2O$ are 2.5×10^{-13} and 2.4×10^{-11}, respectively [37]. The extremely low Ksp values indicate that the reaction products have excellent chemical stability and high bonding potential. Pb could be chemically stabilized through the precipitation of lead phosphate and pyromorphite in the crystal lattice of MPC, resulting in low leachability [38]. The literature reported that 40% MSWI FA blending could be immobilized by MPC, while only 20% MSWI FA replacement was suitable as a component of MPC-based construction applications [39]. At the same dosage, the immobilization efficiency of MPC was 18-fold that of OPC [40]. However, MPC applied for S/S has several drawbacks, such as high cost of raw materials and unstable preparation processes [37].

Magnesium oxysulfate cement (MOSC) is composed of MgO and $MgSO_4$, and is an innovative cementitious material with low carbon emissions [31]. Compared to Ca-based binders, the MOSC system has better compatibility with zinc by avoiding the formation of complexation between Ca^{2+} and $Zn(OH)_3^-$ [41]. In addition, the MOSC system showed excellent stabilization ability for Zn, which is one of the major PTEs in MSWI FA. These characteristics indicate that the MOSC cementitious system could incorporate a large amount of MSWI FA. The results showed that a 33 wt% tailored MOSC binder could achieve satisfying performance for MSWI FA S/S products [42].

Alkali-activated cement

Alkali-activated cement (AAC) is a kind of low-carbon binder, which could be composed of waste materials with pozzolanic activity or latent hydraulicity [43]. During the alkali-activated reaction, the amorphous three-dimensional network structure formed via the dissolution of aluminosilicate in alkaline solutions [44]. AAC shows superior durability to acids, sulfates, and high temperatures compared to OPC, which could serve as a promising binder for the S/S of MSWI FA [45, 46]. Meanwhile, chemical adsorption of dissolved PTEs through electrostatic attraction occurs on the negatively charged hydration gels [47]. However, the PTE leaching behavior is strongly affected by pH [48], which means that the long-term stability of the AAC binder requires further investigation.

In summary, the PTEs in MSWI FA could be immobilized in various cement-based matrices. However, there are several concerns about achieving a green and safe strategy. A large amount of MSWI FA would impede the hydration process and weaken the strength development of the S/S block due to the high contents of pollutants and impurities. On the other hand, the long-term stability of immobilized PTEs should be monitored and improved.

Immobilization mechanisms of PTEs in the S/S matrix

Chemical adsorption, co-precipitation, cations replacement, and physical encapsulation could be the major PTE immobilization mechanisms in cement matrices. PTEs in MSWI FA could interfere with the hydration process. The immobilization mechanisms of different PTEs are relatively distinctive. The characterization of micro-morphology and chemical compositions in S/S blocks is important to speculate on the immobilization mechanism. On the other hand, the effect of PTEs on the hydration reaction in various binder systems should be investigated solely for a detailed explanation.

C–S–H gel owns a large reactive surface area and high-density hydrogen bonding net, which provide large amounts of binding sites for PTEs. Rich Ca in C–S–H provides positive charges for anion stabilization, such as Cl^- and SO_4^{2-}, while the C–S–H gel with silicon is preferential to adsorb cations [49]. The chemical adsorption of PTEs could be dramatically affected by pH value in the matrices. A weak alkaline environment ($pH<11$) would be more suitable to immobilize most PTEs [50]. In addition, the C–S–H gel has a large cation exchange capacity. Some PTEs, such as Cd^{2+}, Zn^{2+}, and Pb^{2+}, are prone to ion exchange to replace Ca^{2+} due to their stronger adsorption affinity [51]. Moreover, the C–S–H gel could solidify PTEs and organic pollutants in its structure physically.

AFt phase, $3CaO\cdot Al_2O_3\cdot 3CaSO_4\cdot 32H_2O$, is one of the major hydration products of cement, but it can be easily transformed into AFm ($Ca_2Al(OH)_6\cdot 0.5\ \times\cdot H_2O$) [14] when the sulfate content is insufficient. AFt could adsorb and immobilize the PTEs via the isomorphous replacement and chemical adsorption. Research has shown that AFt has a very strong binding effect on PTEs. The divalent metals (Pb^{2+}, Zn^{2+}, Ni^{2+}, and Cd^{2+}) could replace Ca^{2+} in AFt phase, while the trivalent metals (Cr^{3+}) could replace Al^{3+} [52]. On the other hand, toxic anions (Cl^-, CO_3^{2-}, SeO_4^{2-}, BrO_3^-, AsO_2^-, and ClO_3^-) could also be embedded in the lattice to replace SO_4^{2-}[53]. AFm has a similar ion exchange capacity with a more stable structure. Isomorphous replacement as the major immobilization mechanism occurred in AFt and AFm that would not change their crystal structure, and the chemical bonding formation during solidification would be insusceptible to varying environmental pH values.

In summary, the C–S–H gel provides a large surface area with chemical binding sites for adsorption of PTEs via electrostatic attraction and ion exchange, and it could also immobilize PTEs in the structure in a physical way. AFt and AFm offer a regular and stable crystal lattice for PTEs isomorphous replacement and physical encapsulation.

Perspectives

Based on the literature review, the strategies for the S/S of MSWI FA are provided as follows: (i) Comprehensive characterization and measurement of the MSWI FA is essential for the S/S binder selection. The chloride, sulfate, and PTEs contents have significant effects on the hydration reaction. Various MSWI FA characteristics also determined the leaching behaviors of PTEs. (ii) The role of the hydration products in the S/S of PTEs in MSWI FA requires further investigation, especially for novel binder systems. The difference between Ca- and Mg-based cements on the PTEs immobilization performance and mechanisms should be explored through advanced characterization. (iii) Comprehensive and integrated leaching procedures should be developed to stimulate the long-term stability of the S/S system under aggressive environmental conditions. An evaluation framework, as well as a machine learning method, should be constructed for designing S/S technologies. (iv) Cost–benefit analysis and life cycle assessment should be conducted to evaluate the economic efficiency and environmental impacts of various S/S processes. Turning hazardous MSWI FA into eco-friendly construction materials is a promising strategy, that fits well with global Sustainable Development Goals (SDGs).

Conclusions

This article introduces the major characteristics of MSWI FA and the low-carbon S/S technologies for MSWI FA treatment. Co-precipitation, chemical adsorption, ion replacement and physical encapsulation are the major mechanisms for the S/S of PTEs. The physicochemical property of MSWI FA and characteristics of binders jointly determined the S/S performance. Novel low-carbon binders should be tailored to enhance the compatibility with various PTEs in MSWI FA. Moreover, the future prospects were proposed for developing new-generation S/S technologies for MSWI treatment in a sustainable way.

Acknowledgements The authors gratefully acknowledge the financial support from the Open Project of State Key Laboratory of Clean Energy Utilization, Zhejiang University (No. ZJUCEU2022001) for this study.

Declaration

Conflict of interest Jianhua Yan is the Editor-in-Chief of Waste Disposal & Sustainable Energy, Qunxing Huang is the Editorial Board member of Waste Disposal & Sustainable Energy. The authors declared that they had no conflicts of interest to this work.

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