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Hydration characteristics and modeling of ternary system of municipal solid wastes incineration fly ash-blast furnace slag-cement

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HIGHLIGHTS

- MSWI fly ash can activate GGBFS through sulfates and chlorides.
- Maximum of AFt and Cl-AFm is obtained by hydration of 55% MSWI FA and 45% GGBFS.
- Cr and Cr(VI) in MSWI fly ash can be stabilized by Cl-AFm and ettringite.

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ABSTRACT

Municipal solid waste incineration fly ash (MSWI FA) can be reused as an admixture in preparing various types of cementitious materials. In this work, MSWI FA was used as a supplementary cementitious material in combination with ordinary Portland cement and ground-granulated blast furnace slag (GGBFS). Mixture design modeling and Gibbs Energy Minimization Software (GEMS) were adopted to discuss the compressive strength and hydration characteristics of binary and ternary systems, respectively. A toxicity characteristic leaching procedure test of total Cr and Cr(VI) was conducted to analyze the environmental risk of MSWI FA-containing samples. The experimental data and the results of mixture design modeling suggested that, with an increase in MSWI FA in GGBFS-MSWI FA binary, the compressive strength exhibited an inverted 'V' shape. GGBFS could be activated by chloride and sulfate in MSWI FA to form ettringite and Friedel's salt in the GGBFS-MSWI FA binary system, according to the results of GEMS and X-ray diffraction. At nearly 0.55 of MSWI FA proportion in the GGBFS-MSWI FA system, the compressive strength and the modeling volume of ettringite and Friedel's salt reached their maximum. The concentration of leached Cr and Cr(VI) indicated that the hydrates from the solidified mixtures could reduce the leaching rate of total Cr and Cr(VI).

1. Introduction

Incineration can reduce municipal solid wastes by more than 80% and is thus widely used as an effective technology for managing municipal solid wastes. However, municipal solid waste incineration fly ash (MSWI FA), as an incineration residue, typically contains high proportions of heavy metals and dioxins and should therefore be treated prior to final disposal to reduce the potential leaching of contaminants [1]. The treatment methods of MSWI FA include (1) taking pretreatments and landfill as hazardous waste, (2) solidification and stabilization (S/S), and (3) separating heavy metals and fly ash (FA) and applying appropriate treatments to both. The landfill method of treating MSWI FA is widely used in China [2]. However, its use is limited because of the high overall costs of landfill sites, stringent regulations, diminishing land availability, and widespread public opposition to the siting of new landfills [3].

Industrial wastes, such as granulated blast-furnace slag (GGBFS) and coal FA, are widely adopted as complementary cementitious materials. MSWI FA shares certain characteristics of GGBFS and FA (such as chemical composition and heat history) but is inferior in performance [4]. The use of MSWI FA as a complementary cementitious material is a promising treatment method considering environmental concerns and technical support. A few studies

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have used MSWI FA to produce different kinds of cementitious materials, such as Portland [5–7], sulfoaluminate [8], alinite cement [9], and intermediate-calcium cementitious materials [10]. This application can resolve the detoxification and disposal problems of MSWI FA. However, many studies [11–13] have shown that the hydration reaction activity of MSWI FA is low. Mixing MSWI FA delays cement hydration; only when the amount of added MSWI FA is large enough can cement mortar strength be significantly reduced [4]. A large amount of MSWI FA in cementitious materials has potential risks to the environment. A recent research [10,14] found that a new hydration phase of ettringite and Friedel's salt formed in a GGBFS-MSWI FA matrix. Thus, the compressive strengths and immobilization effective of heavy metals increased compared with that of a pure ordinary Portland cement (OPC)-MSWI FA matrix. However, excessive MSWI FA in a GGBFS-MSWI FA system can still lead to environmental risk.

This work aimed to use a supplementary cementitious material in combination with OPC and GGBFS, explore the ternary system of OPC-MSWI FA-GGBFS, and determine the optimum proportion of MSWI FA on the properties of compressive strength and Cr immobilization. The optimum proportion of MSWI FA with respect to the properties of cementitious materials (for example, compressive strength) was determined by a statistics-based mixture design model [15,16]. The cementitious characteristics and hydration products were modeled using the Gibbs free energy minimization program GEM-Selektor (GEMS) [17,18] together with thermodynamic data from the Paul Scherrer Institute (PSI)/GEMS database at 25 °C and 1 bar pressure. In addition, a toxicity characteristic leaching procedure (TCLP) test of total Cr and Cr(VI) was conducted to analyze the environmental risk of MSWI FA-containing samples.

Table 2
Physical properties of raw materials.

Physical properties	MSWI FA	GGBFS	OPC
Specific gravity	1.47	2.97	3.21
Specific surface, Blaine, m^2/g	204	434	360
80 μm sieve passing percentage, %	61.55	95.75	92.31
45 μm sieve passing percentage, %	23.76	80.21	75.48

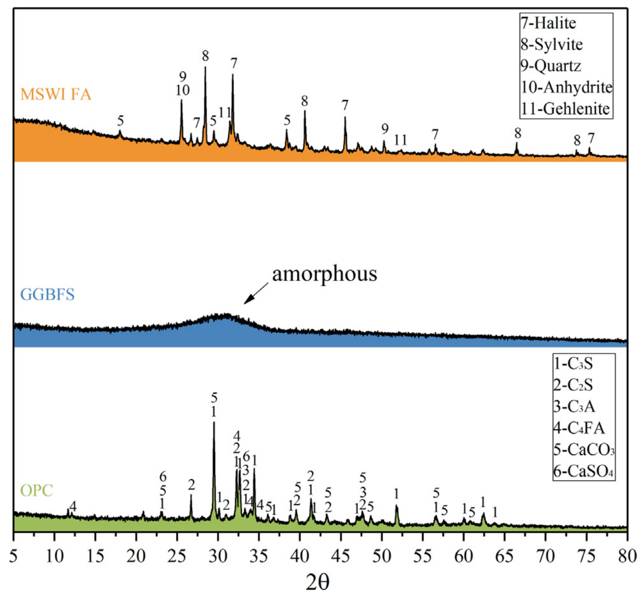

Fig. 1. XRD spectra of MSWI FA, GGBFS, and OPC.

2. Materials and methods

2.1. Materials

MSWI FA, GGBFS, and OPC 42.5 were used in this study for a ternary solidification/stabilization (S/S) cementitious system. The MSWI FA was collected from a municipal waste incineration plant in Guangzhou, China. The GGBFS (S95) was collected from Wuxin Materials Company (Wuhan, China). OPC 42.5 was supplied by Huaxin Cement Corporation (Wuhan, China). The chemical compositions of these raw materials were determined by an X-ray fluorescence spectrometer (XRF) (Bruker AXS, GmbH, Germany), and the trace element contents are presented in Table 1. The physical characteristics of the raw materials are given in Table 2. The TCLP leaching concentrations of Na, K, and Cl of MSWI FA were 3.58, 1.71, and 6.35 mg/L, respectively.

The X-ray diffraction (XRD) (D8 Advance, Bruker AXS, Germany) analysis results of the raw materials are shown in Fig. 1. The major phases of OPC were $3CaO{\cdot}SiO_2$ (C_3S), $2CaO{\cdot}SiO_2$ (C_2S), $3CaO{\cdot}Al_2O_3$ (C_3A), $4CaO{\cdot}Al_2O_3{\cdot}Fe_2O_3$ (C_4AF), gypsum, and calcite. The mineralogical phases of MSWI FA were mainly composed of $CaSO_4$ (anhydrite), KCl (sylvite), NaCl (halite), SiO_2 (quartz), $CaCO_3$, and $2CaO{\cdot}Al_2O_3{\cdot}SiO_2$ (gehlenite). GGBFS exhibited an amorphous structure. The scanning electron microscopy (SEM) (Hitachi S-4800, Japan) images of the raw materials are shown in Fig. 2. The size of agglomerated MSWI FA particles was larger than those of OPC and GGBFS.

2.2. Mixture design process

The relative significance of main mixture parameters and their bonded effects on cementitious material systems are typically analyzed using statistical modeling approaches. A mixture design approach was used in the present work. Unlike factors in classical factorial and response surface designs, those in a mixture design must always total 1. Thus, these factors are not independent [19,20]. A simplex lattice mixture design with three factors and five levels was utilized to evaluate the effect of three types of binding materials (OPC, GGBFS, and MSWI FA) on the properties of an S/S cementitious system. The simplex lattice design is a space-filling design that creates a triangular grid of combinations, as shown in Fig. 3, where the number of combinations (C) is as follows:

$$N = \frac{(m+n-1)!}{n!(m-1)!} \quad (1)$$

where m is the number of ingredients in a mixture and n is the number of levels. When m is given, the number of combinations is determined by the value of n. In this study, the ingredients in the mixture were OPC, GGBFS, and MSWI FA. The sides of the triangle in Fig. 3 were divided equally into 5. In other words, m and n are 3 and 5, respectively. The number of combinations is 21, according to Eq. (1).

With the application of this approach, a mathematical model used to describe the effect of OPC, GGBFS, and MSWI FA proportions and blends on given material system properties was established. To satisfy the accuracy of prediction and avoid overfitting, the regression model of the simplex lattice design was set to be no higher than 4th order [21]. In this study, a second-degree model was used according to three non-independent variables (proportions of OPC, GGBFS, and MSWI FA) and five levels. The model was simplified and listed as follows:

$$Rc = b_1 \times \text{OPC} + b_2 \times \text{GGBFS} + b_3 \times \text{MSWI FA} + b_{12} \times (\text{OPC} \times \text{GGBFS}) + b_{13} \times (\text{OPC} \times \text{MSWI FA}) + b_{23} \times (\text{GGBFS} \times \text{MSWI FA}) \quad (2)$$

Table 1
Chemical composition and trace elements of raw materials.

Analysis (wt%)	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	Cl	K_2O	CaO	Fe_2O_3	LoI
MSWI FA	10.0	1.7	2.4	7.1	1.10	10.4	19.9	6.1	31.9	1.4	4.8
GGBFS	0.4	8.1	16.7	32.2	0.1	2.8	0.1	0.6	37.9	0.3	0.6
OPC	0.3	1.6	5.4	21.5	0.0	0.9	0.0	0.3	61.5	3.9	2.3
Trace elements (mg/kg)	Cd	Cr	Cu	Pb	Zn						
MSWI FA	231.2	2793.1	689.7	1397.3	6917.4						
GGBFS	21.7	79.3	135.5	80.3	17.4						
OPC	106.3	93.8	83.9	65.1	124.7						