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Solidification and immobilization of MSWI fly ash through aluminate geopolymerization: Based on partial charge model analysis

Lei Zheng^{a,b}, Wei Wang^{c,d}, Xingbao Gao^{e,*}

^a School of Energy and Environmental Engineering, University of Science and Technology Beijing, Beijing 100083, China

^b Beijing Key Laboratory of Resource-oriented Treatment of Industrial Pollutants, China

^c School of Environment, Tsinghua University, Beijing 100084, China

^d Key Laboratory for Solid Waste Management and Environment Safety (Tsinghua University), Ministry of Education of China, Beijing 100084, China

^e State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

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ABSTRACT

This study presents an integrated synopsis of the solidification and immobilization mechanisms of fly ash-based geopolymers. A rational analysis of the ion reactions involved in geopolymerization was conducted using the partial charge model (PCM). The following conclusions were obtained: (1) heavy metal cations cannot be immobilized as counter cations through exchange with Na^+ ; (2) isomorphous substitution of heavy metals in the geopolymer can be expected from the condensation reaction between the hydrolyzed heavy metal species and aluminosilicate; (3) the hydrolyzed species condensation could result in solidification and immobilization and be promoted by aluminates; and (4) a geopolymer with the highest immobilization and solidification efficiency can be obtained at an intermediate pH value. The partial charges on the framework of Si, Al, and O in the primary building blocks of aluminosilicate and heavy metal-doped aluminosilicate were confirmed through XPS and ^{29}Si NMR spectroscopy analyses. The effects of activator dosage and types on fly ash-based geopolymers were also investigated, and the results verify the PCM analysis. A geopolymer with the highest strength was produced at an intermediate alkaline dosage. Silicate or aluminate introduced into the activator improved the strength and immobilization efficiency, and aluminate exhibited better performance. Heavy metals bound to the exchangeable or acid-soluble fraction were transformed into aluminosilicate species during geopolymerization.

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1. Introduction

Municipal solid waste incineration (MSWI) fly ash, which contains heavy metals, is classified as hazardous waste in many countries and should be disposed in secured landfills. The disposal of MSWI fly ash should satisfy the following conditions: (1) powerful fixing capacity for heavy metals, (2) structural stability of the product with comparable strength, and (3) low volume-added rate. Traditional stabilization technologies for treatment of heavy metal wastes include cement stabilization/solidification, lime stabilization, and coal ash stabilization. These technologies could result in volume increase using cement or lime. Chemical stabilization and landfill or reuse are recommended because they are environmentally safe and cost effective (Colangelo et al., 2015); these techniques

could also satisfy the requirements for heavy metal treatment. Moreover, the solidification and immobilization of MSWI fly ash through geopolymerization could satisfy the above requirements (Zheng et al., 2011).

Geopolymers (Feng et al., 2012) are framework structures produced by condensation of tetrahedral aluminosilicate units. Tetrahedral frameworks are linked by shared oxygen as poly(sialates) or poly(sialate-siloxo) or poly(sialate-disiloxo) depending on $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio in the system, with alkali metal ions balancing the charge associated with tetrahedral Al. The connection of the tetrahedral frameworks occurs via long-range covalent bonds. Thus, geopolymer structure is perceived as dense amorphous phase, consisting of semi-crystalline 3D aluminosilicate microstructures. Geopolymerization occurs at ambient or slightly elevated temperature, at which leaching of solid aluminosilicate raw materials in alkaline solutions leads to transfer of leached species from the solid surfaces into a growing gel phase; this process is followed by nucleation and condensation of the gel phase to form a solid binder.

* Corresponding author at: State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China.

E-mail address: gaobx@craes.org.cn (X. Gao).

Discussion on immobilization mechanism relies on the following similarities of geopolymers to zeolites (van Jaarsveld et al., 1997; van Deventer et al., 2007): (1) metal ions are included into the geopolymeric network, (2) metal ions are bound to the structure for charge-balancing roles, and (3) a precipitate containing heavy metals is physically encapsulated. However, careful investigation is required for two reasons: (1) geopolymer and zeolite are different. The main difference is that geopolymers are amorphous, in contrast to zeolite, which is crystalline. TEM analysis shows that the microstructure of geopolymers at the nanometer scale comprises small aluminosilicate clusters with pores dispersed within a highly porous network. The cluster sizes are between 5 and 10 nm (Huang et al., 2011). (2) Geopolymerization technology immobilizes heavy metals in the original material during geopolymer synthesis, whereas zeolite is commonly synthesized first and used to adsorb heavy metal ions. Therefore, the precise exact mechanism through which heavy metal immobilization occurs is not yet fully understood in geopolymerization technology.

Studies have investigated the role of synthesis parameters, such as alkaline activator dosage, activator type, and Si/Al ratio, on solidification and immobilization efficiency (van Jaarsveld et al., 1997; Phair et al., 2004). Subsequent research has focused on geopolymers contaminated with different heavy metal salt forms, such nitrate, chromate, and other sparingly soluble salts (Zhang et al., 2008). In addition, leaching tests have been used to evaluate the release behavior of heavy metals from geopolymers and elucidate the immobilization mechanism (Phair and Van Deventer, 2001; Bankowski et al., 2004). Despite these developments, the heavy metal reaction pathway during geopolymerization has been rarely investigated.

The primary chemical processes involved in geopolymerization include particle deconstruction (hydrolysis) and polymerization (condensation) (Weng and Sagoe-crentsil, 2007; Yao et al., 2009; Rozineide et al., 2013); rational analysis of these reactions for silicon and alumina components was previously conducted using the partial charge model (PCM) (Weng et al., 2005; Weng and Sagoe-crentsil, 2007). In the present paper, this model was adopted for MSWI fly ash solidification and immobilization during geopolymerization through qualitative and quantitative analyses of the PCM results. This model can be used to elucidate the reaction route of heavy metal cations and major cations of MSWI fly ash during geopolymerization and provides an enhanced understanding of solidification and immobilization mechanisms. This paper presents a description of the main chemical processes of cations involved in aqueous chemistry during geopolymerization by using PCM. The PCM results are verified through comparison with experimental data (XPS and MAS NMR spectroscopy).

According to the PCM analysis, synthesis parameters in the geopolymerization of MSWI fly ash were optimized in the present study. Previous studies used hydroxide, silicate, or their mixture as the main activator in geopolymerization. Sodium aluminate has also been used as an activator in geopolymers (Phair et al., 2002; Hajimohammadi et al., 2008, 2010). However, systematic evaluation of solidification and immobilization during geopolymerization using aluminate activator, with focus on reaction mechanisms, is limited. In the present study, aluminate was used to synthesize MSWI fly ash-based geopolymers. The effects of alkaline activator dosage and types on the compressive strength, microstructure, and heavy metal immobilization efficiency of geopolymers were determined and correlated with the PCM analysis results.

This study provides an integrated synopsis of the stabilization behavior of MSWI fly ash-based geopolymers using aluminate as well as the solidification and immobilization mechanisms of MSWI fly ash during geopolymerization.

2. Material and methods

2.1. Materials

Fly ash was sampled from a MSW incineration plant, which has three sets of 350 t d⁻¹ stoker furnace, in East China. Each furnace contains a semi-dry lime scrubbing system and fabric bag filter precipitator. Major element analysis was performed by X-ray fluorescence spectrometry (XRF, Shimadzu Lab Center XRF-1700, Japan). The concentration of trace elements was analyzed after the sample was microwave-digested with acid according to a previously published protocol (Mester et al., 1999). Element concentrations were determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES) using standard addition method (ICP-AES) (Perkin2Elmer Elan 6000, USA). Metakaolin (MK, kaolinite calcined at 650 °C) was used to prepare the geopolymer and characterized by XRF. Results are shown in Table 1. Distilled water and analytical grade NaOH, NaAlO₂, and Na₂SiO₃ were used in all experiments.

2.2. Synthesis of geopolymer

NaOH (15 M), NaAlO₂ (2 M), and Na₂SiO₃ (2 M) solutions were prepared, sealed to avoid carbonation, and incubated at room temperature overnight. The MSWI fly ash or MK (12 g) was first mixed with designed amount of alkali activator. The water-to-solid mass ratio of all batches was fixed at 0.3. According to our preliminary study, this ratio should be higher than 0.18 to ensure easy demolding but should be less than 0.35 to prevent water exclusion during uniaxial pressing (details will be provided in later section). Thus, 0.3 was used in the present study. Cylinder test specimens (U 20 mm and H 40 mm) were formed by uniaxial pressing (6 MPa). The demolded specimens were cured at room temperature prior to analysis.

According to our previous study (Zheng et al., 2011), all tests were performed at 14 d. Table 2 presents a summary of the compositions of all matrices. The Na⁺ dosage in the activator solution was contributed by both NaOH and Na₂SiO₃ or NaAlO₂, and the Si or Al dosage was accounted only from Na₂SiO₃ or NaAlO₂. Series 1 was performed to investigate the influence of alkaline activator dosage on the strength of the fly ash-based geopolymer with fixed amount of Al added (0.2 mol kg⁻¹). Series 2 was designed to investigate the effect of aluminate or silicate addition on the strength, microstructure, and heavy metal immobilization capability of fly ash-based geopolymers with fixed Na⁺ dosage (2.5 mol kg⁻¹). In Series 2, “G-FA-M-b” (where “M” is “Si” or “Al,” which indicates that silicate or aluminate is used in the activator, and “b” is a number) was

Table 1
Chemical composition of calcined kaoline and MSWI fly ash.

Calcined kaoline compositions			
Major components	SiO ₂	Al ₂ O ₃	others
Value (wt%)	53.1	45.3	1.6
MSWI fly ash compositions			
Major components	Value wt%	Trace elements	Value mg kg ⁻¹
CaO	25.2	Zn	5692
Cl	18.7	Cu	3315
SiO ₂	15.9	Pb	933
SO ₃	9.1	Cd	423
Al ₂ O ₃	8.9		
Na ₂ O	7.2		
K ₂ O	5.7		
Fe ₂ O ₃	3.5		
MgO	2.9		
Others	2.9		

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