



# Effect of blast furnace slag grades on fly ash based geopolymer waste forms



Hui Xu, Weiliang Gong\*, Larry Syltebo, Kevin Izzo, Werner Lutze, Ian L. Pegg

Vitreous State Laboratory, The Catholic University of America, 620 Michigan Ave NE, Washington, DC 20064, USA

## HIGHLIGHTS

- Slags of grades 80, 100 and 120 investigated on fly ash based geopolymer waste forms.
- Concentrated Hanford radioactive waste simulant consisting of more than 20 chemicals.
- Higher grade slag not definitely leading to higher strength or shorter setting times.
- More hydration heat for higher grade slag confirmed by calorimetric study.
- Reactivity of higher grade slag found to be better exploited at enhanced Si/Ca ratios.

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## ABSTRACT

Ground granulated blast furnace slags (GGBFSs) of grades 80, 100 and 120 were investigated for high waste loading fly ash based geopolymer waste forms. Samples were prepared at a fixed fly ash/GGBFS mass ratio of 5/3, using an activating solution prepared from concentrated Hanford secondary waste (HSW) simulant. The fresh pastes were subjected to isothermal conduction calorimetry and Vicat setting time measurements, and the cured waste forms were characterized by compressive strength test, XRD and SEM/EDS analyses, as well as the TCLP leaching test. The results show that GGBFS of higher grade generated more hydration heat, yet not definitely led to higher compressive strength or shorter setting times, suggesting that the GGBFS grading index established for cement industry may not be simply introduced to geopolymer application. It was also found that the reactivity potential of high grade GGBFS in fly ash based geopolymer might be better exploited at enhanced  $\text{SiO}_2/\text{Al}_2\text{O}_3$  and  $\text{SiO}_2/\text{CaO}$  ratios. Except for rhenium, which is not regulated in TCLP, all heavy metals and hazardous elements in the HSW simulant were effectively immobilized by the geopolymer waste forms. However, effect of different GGBFS on heavy metals and hazardous elements fixation depended on different metals and elements.

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## 1. Introduction

Geopolymers are a class of synthetic alkali-activated aluminosilicate inorganic polymers (AIPs) featuring a predominantly X-ray amorphous three-dimensional network [1]. The term geopolymer was coined by Joseph Davidovits three decades ago [2].

*Abbreviations:* GGBFS, ground granulated blast furnace slag; HSW, Hanford secondary waste; AIPs, aluminosilicate inorganic polymers; C–A–S–H, aluminium-modified calcium silicate hydrate; N–A–S–H, sodium aluminosilicate hydrate; OPC, ordinary Portland cement; ILAW, immobilized low-activity waste; HEPA, high-efficiency particulate air; PNNL, Pacific Northwest National Laboratory; ICC, Isothermal Conduction Calorimeter; ANSI/ANS, American National Standards Institute/American Nuclear Society.

\* Corresponding author. Tel.: +1 (202) 319 5393; fax: +1 (202) 319 4469.

E-mail address: [gongw@vsl.cua.edu](mailto:gongw@vsl.cua.edu) (W. Gong).

As described by Davidovits, the fundamental structure of geopolymers consists of  $\text{SiO}_4$  and  $\text{AlO}_4$  tetrahedrons linked alternatively ( $-\text{SiO}_4-\text{AlO}_4-$ , or  $-\text{SiO}_4-\text{AlO}_4-\text{SiO}_4-$ , or  $-\text{SiO}_4-\text{AlO}_4-\text{SiO}_4-\text{SiO}_4-$ ) by sharing all oxygen atoms between two tetrahedral units. The negative charges on tetrahedral  $\text{AlO}_4$  are balanced by alkalis cations (typically  $\text{Na}^+$  and/or  $\text{K}^+$ ) provided by alkaline activating solution. Theoretically, any aluminosilicate material can be used for geopolymer synthesis.

The geopolymerization of some industrial wastes such as coal ashes has attracted increasing interests since the past two decades [3–7]. Fly ash is generated during combustion of coal in thermal power plants and essentially contains  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  along with other compounds, including  $\text{CaO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{MnO}$ , etc. [8,9]. Fly ash has become an important raw binder material for geopolymer due to its high  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  contents, wide availability in large

quantities, sound property of the hardened products, low cost in comparison to those of traditional calcined natural clays, as well as significant environmental benefits including natural resource preservation, low energy consumption and reduced CO<sub>2</sub> emission [10–13]. However, former studies have also revealed that, generally, the dissolution of fly ash at room temperature had not completed before the final hardened structure was formed [5,6,14]. The relatively low reactivity of fly ash usually led to slow setting of the fresh paste and poor compressive strength of the hardened material [15,16]. In many cases, for fly ash based geopolymers, an elevated curing temperature of 40–85 °C is needed to favor a better compressive strength gain [17,18].

In order to compensate the disadvantages of fly ash in geopolymerization, ground granulated blast furnace slag (GGBFS) has been incorporated into fly ash based geopolymers [19,20]. GGBFS is an amorphous by-product of the steel industry with a latent hydraulic reactivity, which can be catalyzed by proper activators to form cementitious materials [21,22]. The incorporation of calcium-rich GGBFS into fly ash based geopolymers may improve the setting time and compressive strength of the geopolymer by forming aluminium-modified calcium silicate hydrate (C–A–S–H) gel in addition to the sodium aluminosilicate hydrate (N–A–S–H) gel (geopolymer gel) [23,24] and compacting the microstructure [25].

On the other hand, GGBFS has been recognized as one of the major cementitious materials in ordinary Portland cement (OPC) concretes [26]. For the cement industry, an ASTM C989 standard has been established, which classifies GGBFS into three strength grades, i.e. 80, 100 and 120 [27]. According to ASTM C989, the strength grades of GGBFS are determined by their slag activity index

Slag activity index, % =  $SP/P \times 100$

where SP represents average compressive strength of the slag-reference cement cubes consisting of 50 wt% slag and 50 wt% Portland cement, and P is the average compressive strength of the reference cement cubes of 100 wt% Portland cement. The ASTM C989 standard is briefed in Table 1.

Although incorporation of different amounts of one GGBFS into fly ash and/or metakaolin based geopolymers has received intensive investigation [19,20,25,28], the implication of the GGBFS grading index to geopolymer synthesis remains poorly understood. Besides, to the authors' best knowledge, no study on effect of GGBFSs of different grades on chemical durability of fly ash based geopolymer waste forms has ever been published.

The present study, therefore, investigated the effect of GGBFSs of grades 80, 100 and 120 on fly ash based geopolymer waste forms. A concentrated Hanford secondary waste (HSW) stream simulant containing 5 mol/L sodium and spiked with 100 mg/L NaReO<sub>4</sub> (as an analogue for radionuclide <sup>99</sup>Tc) was employed to prepare the alkaline activating solution. The geopolymer waste forms were cured at room temperature and characterized by isothermal conduction calorimetry, setting time and compressive strength measurements, X-ray diffractography (XRD), scanning electron microscopy (SEM), energy dispersive X-ray spectrum

(EDS), as well as the toxicity characteristic leaching procedure (TCLP) leach test [29].

## 2. Materials and methods

### 2.1. Raw materials

The major raw binder materials used in this study are class F fly ash from Brandon Shores Power Plant provided by Separation Technologies LLC, GGBFS grades 80 and 120 provided by Lafarge North America Inc., and GGBFS grade 100 provided by Holcim US Inc. An amorphous silica fume, obtained from Norchem Inc., was employed at 8.67 wt% of the sum of the fly ash and GGBFS as partial replacement of the fly ash and GGBFS in some samples. A Type 5A 8 × 12 mesh molecular sieve (Zeolite 5A), obtained from Delta Enterprises, was ground to fine powders and used as an additive at 1 wt% of the paste. Chemical compositions of the raw materials as analyzed by X-ray fluorescence (XRF) are given in Table 2. Particle size distributions of the fly ash and GGBFSs are shown in Fig. 1.

### 2.2. Hanford secondary waste simulants

Liquid Hanford secondary waste (HSW) simulant S1 was used to prepare the alkaline activating solution for the fly ash based geopolymer waste forms. The HSW simulant S1 represents the baseline for the immobilized low-activity waste (ILAW) off-gas caustic scrubber effluent downstream of the high-efficiency particulate air (HEPA) filters at the Hanford Site, WA. The composition of HSW simulant S1, as provided by Pacific Northwest National Laboratory (PNNL), indicates a 1 mol/L sodium concentration [30]. In this study, the sodium concentration for HSW simulant S1 was increased to 5 mol/L. It is assumed that radionuclide technetium-99 will occur in the HSW waste stream in its most soluble form, i.e. pertechnetate TcO<sub>4</sub><sup>-</sup> [30]. Therefore, rhenium was spiked as NaReO<sub>4</sub> into the simulant as an analogue for technetium-99 in its soluble form <sup>99</sup>TcO<sub>4</sub><sup>-</sup>. The recipe for a 10 L batch of the HSW simulant S1 prepared in this study is presented in Table 3.

### 2.3. Geopolymer preparation

The activating solution was prepared from the HSW simulant S1 without additional water. Under mechanical stirring, sodium hydroxide was dissolved into the simulant. After cooled to room temperature, the solution was mixed with solid binder materials on a Lancaster K-Lab mixer for several minutes. Most of the obtained paste was cast into 5.08 × 10.16 cm cylindrical plastic molds, whereas a small portion of the fresh paste was subjected to the setting time test. The molded samples were immediately placed on a vibrating table for about two minutes to remove entrapped air bubbles. Then, the samples were sealed with lids and cured at room temperature for designated ages.

For all the GGBFSs, two formulations, A and B, were designed to prepare the fly ash based geopolymer waste forms. While formulation A is a simple fly ash-GGBFS system containing 1.0 wt% additive of zeolite 5A, formulation B replaces 8.67 wt% of the sum of the fly ash and GGBFS by soluble silica fume. However, the fly ash/GGBFS mass ratios were kept constant at 5/3 throughout the study. A summarized recipe for the geopolymer waste forms of the two formulations is presented in Table 4. Each recipe was tailored for a 6.2 kg batch of fresh geopolymer paste, usually yielding fourteen to fifteen cylindrical samples.

The nomenclature used for geopolymer sample ID is given in Table 5.

**Table 1**

ASTM C 989 standard for the classification of different grade slag.

Day index	Grade type	Minimum slag activity index%	
		Average of last five consecutive samples	Any individual sample
7 days	Grade 80	–	–
	Grade 100	75	70
	Grade 120	95	90
28 days	Grade 80	75	70
	Grade 100	95	90
	Grade 120	115	110

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