



Increasing mechanical strength and acid resistance of geopolymers by incorporating different siliceous materials

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HIGHLIGHTS

- Sulphuric acid resistance of fly ash/slag based geopolymers are studied.
- Hollow glass microsphere waste is effective of increasing acid resistance.
- Binders with 5% siliceous materials exhibited a more compact microstructure.
- High temperature is beneficial to activate siliceous fillers in geopolymer cement.

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ABSTRACT

This study, for the first time, reports using hollow glass microsphere waste (HGMW) and quartz powder (QP) to improve compressive strength and sulphuric acid resistance of fly ash/slag based geopolymers (AAFS), which is aimed for oil-gas well cementing and other uses where acid corrosion medium exists. Due to the lower reactivity of HGMW and QP compared to fly ash and slag, their incorporation needs a high temperature curing process (at 80 °C in this study) to obtain a certain extent of reaction. The resulting product with 5% HGMW and 5% QP exhibits higher compressive strength than the neat AAFS. After sulphuric exposure for 90 days, the high temperature cured samples show less susceptibility to the acid attack in comparison with those samples cured at room temperature and those without HGMW and QP. It is confirmed that better resistance of acid attack of the 80 °C cured mixture with addition of HGMW and QP than the neat AAFS mixture is due to the more crosslinked and compact microstructure. In addition, the volume stability of the geopolymer binders is also assessed and is found to be satisfying under both the ambient and high temperature conditions.

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1. Introduction

The alkali-activated cements, also broadly called ‘geopolymers’ [1–3], have shown resistance to aggressive chemical corrosion [4,5]. The acid resistance property raises particular interest in the development of oil-gas well cementing materials, which need resistance against SO₂, and other construction materials, such as the industrial wastewater and sewer systems, where acid corrosion is a long-lasting issue. Previous studies have shown that fly ash-

based geopolymers are less susceptible to acid corrosion. Lee and Lee [6] reported that fly ash based geopolymers activated by sodium silicate exhibited higher residual compressive strengths compared to ordinary Portland cement (OPC) binder after immersion in 10% sulphuric acid solution for 90 days. This is due to the substantial difference of the formed gel in chemistry compared to OPC binder, and the low porosity of geopolymer binders. Ariffin et al. [7] compared the performance of blended ash (coal combustion fly ash and palm oil fuel ash) geopolymer concrete and OPC concrete when exposed to 2% sulphuric acid for 18 months. The result indicated the former has lower deterioration rate than the latter, which was attributed to a more crosslinked gel structure formed in the geopolymers. A similar experimental study can also be found in the work of Bakharev [5]. According to literature

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[8–10], the corrosion of the alkali-activated cement and concrete by acid attack involves two processes: (1) ion exchange reactions between sodium and calcium ions as a charge compensating role in gel structure with H^+ and H_3O^+ penetrated from acid solution; (2) electrophilic attack by acid protons on polymeric Si—O—Al bonds.

It has been reported that the availability of silica has an important effect on the process of geopolymerisation, which determines the structure of gel and thus influences the mechanical strength [11], and possibly the acid resistance. Some efforts have been made with attempts to increase the acid resistance in geopolymers. Lloyd et al. [12] studied the effect of soluble silicate content on resistance of sulphuric acid (pH = 1.0) by using a corrosion depth as an index. Their results revealed that an increase of dissolved silicate content in the binder effectively enhanced the acid resistance due to the increase of gel volume and Si/Al ratio in the binder. In addition to soluble silicate, other siliceous additives have also been used, in order to improve the acid resistance. Deb et al. [13] noted that a moderate addition of nano-SiO₂ into fly ash geopolymers has a positive influence on the sulphuric acid resistance. That was attributed to the improved compactness of microstructure and thus impeded ingress of acid into binder. Chindaprasirt et al. [14] reported that an addition of silica fume into fluidized bed combustion fly ash geopolymer can slightly enhance the sulphuric acid resistance, indicated by the residual compressive strength after 3 months of immersion. A more recent study [15] further demonstrated that moderate silica fume substitution can considerably increase the acid resistance of fly ash based geopolymers because of the compactness of binder. Based on the results above, it is hypothesized that theoretically the addition of some siliceous materials into alkali-activated cements is in favor of increasing acid resistance.

Considering the oil-gas well cementing, where high temperature (80–150 °C) and, sometimes, acid environment challenge the long stability of Portland cement (PC), additives that can increase thermal stability of PC are required. Quartz powder (QP) is one of the additives. QP has particle sizes from 0.1 to 100 μm, which can be used in ultra-high performance concrete as inert fillers as well [16,17]. Zhang et al. [18] reported that the compressive strength of Class-G oil well cement with addition of 30% quartz sand increased at 2 days under the temperature between 100 and 150 °C. QP has been attempted in the production of the geopolymers recently. Rashad et al. [19] studied the compressive strength of slag-based geopolymer (AAS) with 0 to 30% QP replacement under both non-autoclaved and autoclaved curing (8 bars, 170 °C). Their results showed that samples with QP addition have higher strength than the neat AAS after 28 days ambient curing, which was attributed to the refined pore structure in matrix by the space filling effect of QP particles. In addition, the strength of samples with QP incorporated under autoclaved curing was higher than the neat AAS samples, which was due to the formation of reaction products during further possible reactions of un-reacted particles under autoclaved condition. By the same researchers [20,21], metakaolin-based geopolymer (AAMK) and fly ash-based geopolymer (AAFA) substituted by 0–30% QP were investigated in terms of changes in the workability and compressive strength development under thermal conditions. The results indicated that the workability increased with the addition of QP and the pastes exhibited better performance than those of the neat AAMK and AAFA, due to the space filling effect of QP particles.

In this study, besides QP, we consider to use a new siliceous material as well, the hollow glass microsphere waste (HGMW). HGMW is a waste material generated in the production of hollow glass microsphere, which is widely used in construction and oil well cementing works for lightening body weight, and increasing heat insulation and corrosion resistance [22,23]. In terms of HGM

production in China, the common processes include spray drying, high temperature heat treatment, with sodium silicate and boric acid used as main raw materials. In this process, million tons of particles that do not meet the quality requirements including sphericity, wall thickness and density, are delimited as a waste, i.e. the HGMW.

There are very limited literatures reporting the application of QP in geopolymer for acid resistance and none in HGMW; the previous works have mainly focused on the high temperature resistance [19–21]. In this study, we aim to utilize the two siliceous materials in the development of oil well cementing materials, which can also be used in constructions as well where acid resistance property is required. For this purpose, setting time and early reaction process of the fresh pastes are analyzed and the compressive strength development of samples cured at 20 °C and 80 °C is studied. The matured samples are further tested for acid attack and are investigated by directly observing image of the samples and compressive strength changes. X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and mercury intrusion porosimetry (MIP) are used to provide a better understanding of the samples microstructure before and after acid attack.

2. Experimental programs

2.1. Materials

The fly ash was obtained from Huaneng power station (Nanjing, China) and the granulated blast furnace slag was supplied by Jiahua Co. Ltd. (Nanjing, China). HGMW and QP were supplied by Bengbu Design and Research Institute of Glass Industry (Anhui, China). The chemical compositions of four materials, as analyzed by X-ray fluorescence (XRF), are shown in Table 1. The main chemical compositions of the HGMW are SiO₂, CaO and Na₂O, and the QP contains 99% SiO₂. Table 2 gives a summary of the physical property for the four materials. Fig. 1 shows the mineralogical compositions of the four materials. The results of XRD analysis show that QP is well crystalline and HGMW is completely amorphous.

The SEM images of the four raw materials are presented in Fig. 2. HGMW particles exhibit a near-spherical morphology. The particle size distributions of the four materials as tested using a laser particle size analyzer and the results are plotted in Fig. 3. The d_{50} and d_{90} of HGMW are 19.0 μm and 20.0 μm; while d_{50} and d_{90} of QP are 35.2 μm and 47.3 μm. The slag and fly ash have smaller particle sizes than the QP and HGMW.

The alkaline activating solution was prepared by dissolving sodium hydroxide pellets (analytical reagent) into the mixtures of sodium silicate solution (Ms = 3.24, 8.21%wt Na₂O, 25.8%wt SiO₂) and water with a weight ratio of 0.14:1:0.35. The modulus and concentration of activator were 1.4 and 30% (mass fraction of SiO₂ and Na₂O) respectively. The activator was cooled to the room temperature at least 3 h prior to use.

2.2. Synthesis of pastes

The mixing proportions of the four materials are shown in Table 3. The ratio of slag to fly ash was fixed at 3–7 parts by weight. The liquid (including water content) to solid ratio (by weight) was set at 0.60, which gives a good flowability. Solid materials were dry mixed for 3 min and then mixed with the activator for 2 min. The pastes were casted into $\Phi 25.4$ mm \times 25.4 mm steel molds for compressive strength test and 20 \times 20 \times 80 mm cuboid molds for linear shrinkage test. All pastes were sealed to cure at 20 \pm 2 °C for 8 h to obtain initial strengths because of the different setting time of mixtures. After demolded, the specimens were divided into two groups: one group of samples were cured at 20 \pm 2 °C in sealed

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