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Original Research Paper

Influence of sepiolite addition on mechanical strength and microstructure of fly ash-metakaolin geopolymer paste

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ABSTRACT

This work aims to verify the feasibility and broaden the application of utilizing acicular microfiber sepiolite to make reinforced geopolymer composites. Compressive strength, flexural strength and microstructure of fly ash-metakaolin (MK) geopolymer activated by sodium silicate and sodium hydroxide solutions blended with 0–20% sepiolite by mass with an interval of 5% were investigated. The experimental results reveal that sepiolite exhibits little effect on compressive strength before 3 days of curing. After 7 days, the addition of up to 10% sepiolite improves compressive strength, however, a further increase in the sepiolite leads to decrease in strength. Similarly, up to 10% sepiolite addition exhibits improving effect on flexural strength regardless of days of curing. With the increasing addition levels of sepiolite up to 10%, compact matrix can be observed compared to the reference geopolymers. Some crack can still be observed in specimen containing 20% sepiolite as compared to specimens containing 10% sepiolite. For geopolymer containing 10% sepiolite, cumulative intrusion volume corresponding to the porosity decreases. Further addition of sepiolite enlarges the cumulative intrusion volume and shifts the most probable pore diameters to higher values. The results from mechanical properties and microstructure observation are compatible. This work indicates that the optimal replacement of sepiolite in geopolymer is not exceeding 15%, and sepiolite is beneficial for compressive and flexural strength as well as pore refinement and denser microstructure. It will broaden the application of sepiolite in geopolymer composites.

1. Introduction

The main drawbacks of cement including annual growing production of Portland cement [1] and the greenhouse effect and global warming accelerated by CO_2 emission from cement manufacturing [2] prompt various researches in an attempt to develop geopolymer. As a new kind of environment friendly inorganic binder, geopolymer was derived by alkali activated aluminosilicate source material (including metakaolin, fly ash, red mud and slag) and was first developed by Davidovits [3]. Geopolymer exhibits comparable mechanical properties and durability characteristics as that of Portland cement, but has lower energy requirements and lower CO_2 emissions during its production [4].

The formation of geopolymer involves the dissolution of aluminosilicate species in an alkaline environment to form an amorphous three-dimensional geopolymer network by polycondensation reaction [5], therefore, it has similar or even better properties compared to ordinary Portland cement such as early compressive strength, low permeability, good chemical resistance and excellent fire resistance behaviour [6–11].

Diverse industrial by-products and aluminosilicate source materials were proven to be suitable for producing geopolymer materials. For example, fly ash, has properties typical of industrial waste that can be found all over the world, it is particularly attractive for the synthesis of geopolymers. The properties of fly ash-based geopolymer have been extensively studied in the last decades [12–16]. The reported findings indicated that fly ash is suitable for making geopolymer and the resulting geopolymer presents high mechanical properties and durability [17–20]. Furthermore, metakaolin with high reactivity has been extensively studied to understand the mechanisms of geopolymer formation

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and was also proven to be suitable for producing geopolymer materials [21] with good mechanical properties [22–24]. Additionally, Duan et al. [24] proposed a novel surface waterproof geopolymer based on alkali activation of metakaolin. This newly developed modification method will broaden the application of geopolymer from environmental and economical consideration, for example, the surface waterproof geopolymer could be used as waterproof layer on marine concrete. Zhang et al. [25] used fly ash to partially substitute metakaolin in geopolymer synthesis, and indicated that the replacement of 10% by fly ash gave an increased reaction extent and also a higher 28-day compressive strength with pore refinement.

As commonly known, cementitious materials are brittle. Traditionally, the most commonly used method to strengthen cementitious materials was to add fibers to make reinforced cementitious materials [26]. In the field of concrete construction, the use of fibers has been steadily increasing over the past years in an effort to overcome the inborn tensile strength and toughness limitations of concrete [27]. Adding fibers into concrete has proven to be an effective method to eliminate its inherent brittleness, producing materials with increased flexural strength, ductility, toughness and improved durability properties [28–34].

However, high cost and health hazard of fibers preparation hinder its application. Sepiolite, formulae ($Si_{12}Mg_8O_{30}(OH)_4(OH_2)_4$), a fibrous clay with thixotropic behavior, is used as a thickener for cement slurry and paint [35]. In concrete aspect, its micro reinforcement features have also been highlighted [36], sepiolite could improve the mechanical properties of concrete.

Sepiolite is a cheap hydrated magnesium silicate mineral belonging to the group of phyllosilicate as well as montmorillonite, kaolin or mica. The main differential characteristics with other members of the group are its acicular morphology. Its acicular morphology can even act as an reinforcement of concrete. Its main industrial uses are in construction, coatings or bitumen industry, along with its applications as absorbent, carrier or binder in a great variety of industries [37]. Results published previously show that the addition of sepiolite to Portland cement mortar reduces the crack width and crack length notably [38].

From the available literature, researchers had conducted tests which showed promising results for sepiolite being suitably used in concrete. Jarabo et al. [39] evaluated the effect of sepiolite on flocculation of fiber-reinforced cement and proved that sepiolite could increase thixotropy of cement slurries for easier processing, to prevent sagging and to provide a better final quality of cement products. Kavas et al. [40] investigated the structural properties and optimum blend ratios of sepiolite-reinforced cement composites and found that the addition of 10% sepiolite fibers enhanced the mechanical and physical properties of the mortar. Martinez-Ramirez et al. [41] observed that sepiolite slowed down the rate of carbonation process in lime mortars due to its capacity for water adsorption without affecting the mechanical behavior of the mortars. Andrejkovicova et al. [42] evaluated the behavior of mortars based on lime with the addition of metakaolin and sepiolite and reported that mortar containing 5 wt.% of sepiolite provided the best characteristics of mortar to be applied as render in the renovation of historical buildings. Fuente et al. [43] demonstrated that the sepiolite could be used in the manufacture of fiber–cement to increase the retention of solids and the drainage rate especially in mixtures containing poly(vinylalcohol) fibers. Melo et al. [44] concluded that the incorporation of siliceous fly ashes in ordinary Portland cement together with addition of MK or/and sepiolite provided the largest expansion speed increase and the lowest relative density of the foamed mortar.

However, after reviewing the previously published findings, we observe that there is a lack of experimental data on sepiolite-added gopolymer. The effects of sepiolite on geopolymers are not well-known, and little information is available about microstructure changes. Quantitative information on effects of sepiolite addition on fly ash-metakaolin geopolymer still requires further investigation.

Therefore, this present study is devoted to determining the mechanical strength, microstructure evolution and pore structure of geopolymer prepared using a combination of fly ash and metakaolin and activated by liquid alkaline activator, with the source material partially replaced by sepiolite at amounts ranging from 0 to 20% (in 5% intervals) by weight. The results of this work will reveal the effects of sepiolite on properties of geopolymer, therefore, after investigating and quantifying the changes in geopolymer properties, it will promote the addition of sepiolite in making geopolymer composites.

2. Experimental process

2.1. Materials

Fly ash utilized in this study was provided by Shenhua Junggar Energy Corporation in Junggar, Inner Mongolia, China. Metakaolin was obtained by calcination of kaolinite from Yunnan, China. As microfibers reinforcement, sepiolite (SP) was also used. The chemical analysis of starting materials mentioned above is listed in Table 1. It indicates that fly ash and metakaolin are rich in SiO_2 and Al_2O_3, which are appropriate to be employed as starting materials for the production of geopolymer by alkali-activated geopolymerization. Furthermore, sepiolite is rich in SiO_2, it changes the Si/Al ratio of the starting materials and leads to the changes of properties of the resulting geopolymer.

The micrographs of fly ash, metakaolin and sepiolite are shown in Fig. 1(a), (b) and (c), respectively.

Scanning electron microscopic study in Fig. 1(a) and (b) reveal some interesting features related to the morphology of fly ash and metakaolin samples. It can be seen that the shape of fly ash is usually irregular and appears to be porous in a potential state of fragmentation with small and rough particles. The microstructure of metakaolin also reveals the irregular and sheet shape. It shows the presence of very fine particles being perceived as an agglomeration of particles. Sepiolite in Fig. 1(c) reveals a acicular morphology.

The particle size analysis of fly ash, metakaolin and SP were also carried out using a laser diffraction particle size analyzer (MASTERSIZER S, Malvern, U.K.). Samples were dried for 24 h at 105 °C in an oven, subsequently each sample was ground and sieved for 30 min in a sieve shaker. The characteristic particle diameters D_3, D_{10}, D_{50}, D_{90}, D_{97} and D_{ave} have been tabulated in Table 2. Different particle sizes are observed in the fly ash, metakaolin and SP. 90% particles in metakaolin are less than 40 μm in size whereas in fly ash, 90%

Table 1
Chemical compositions of starting materials by XRF analysis (mass, %).

	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	MnO	TiO_2	LOI[a]
Fly ash	29.47	51.72	2.25	0.15	5.21	0.05	0.35	0.03	1.83	8.58
Metakaolin	53.32	42.09	2.33	0.21	0.09	0.49	0.64	0.02	0.63	0.08
Sepiolite	36.15	0.76	0.42	17.14	25.03	0.44	0.38	0.02	0.03	19.57

[a] LOI: Loss on ignition.