Construction and Building Materials 170 (2018) 619–626

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Geopolymer synthetized from sludge residue pretreated by the wet alkalinizing method: Compressive strength and immobilization efficiency of heavy metal

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HIGHLIGHTS

- Wet alkalinizing treatment of sludge residue reduced the particle size by 400 times.
- Pretreatment of sludge residue enhanced the compressive strength of geopolymer.
- Content of sludge residue determined the characteristic of geopolymer synthetized.
- Immobilization efficiencies of Cu and Zn in the geopolymer reached more than 95%.

ARTICLE INFO

Article history:
Received 3 November 2017
Received in revised form 6 March 2018
Accepted 7 March 2018
Available online 23 March 2018

Keywords:
Sludge residue
Wet alkalinizing pretreatment
Geopolymer
Compressive strength
Heavy metal immobilization

ABSTRACT

To realize the reclamation of the sludge residue, we used the sludge residue pretreated by the wet alkalinizing method to prepare geopolymer. Results showed that, when the dosage of the alkali activator, the silicate modulus of the alkali activator, and the doping amount of the pretreated sludge residue were 82.5%, 1.3 mol/mol, and 20% respectively, the compressive strength of geopolymer reached a maximum at 89.03 MPa. The preparation of geopolymer also realized a highly-efficient immobilization of the unstable-forms of copper and zinc (72.22% and 68.60%) in sludge residue, which significantly reduced the pollution risks of the leaching of heavy metals.

1. Introduction

The sludge residue generated during sewage treatment is rich in poisonous bacteria, degradation-resistant organic matters, heavy metals, and other pollutants that are harmful to the environment. Bioorganic matter contained in the sludge residue, however, is regarded as a precious resource that could replace fossil fuels if they can be reclaimed efficiently, thereby easing the energy crisis [1]. With increasing quantities of sewage treated around the world, the output of sludge residue inevitably will increase. To date, the annual production of sludge residue (dry basis) in America and Europe has reached 7 and 10 million tons, respectively, whereas production in China has increased to between 60 and 90 million tons. In response to this increased production, people have been exploring efficient treatment methods that could achieve the safe reduction and reclamation of sludge residue. Currently, common sludge residue treatment methods include landfill, agricultural utilization, incineration, and pyrolysis, however, these methods pose certain problems—for example, landfill causes land resource shortages, agricultural utilization results in the heavy metal pollution in soil, incineration produces air pollution with dioxin (a secondary pollutant caused therefrom), and pyrolysis brings with it high energy consumption [2,3]. Under such a backdrop, people have begun to explore new methods to treat the sludge residue. For example, synthetizing the sludge residue into new industrial building material not only realizes the reduction and reclamation of

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https://doi.org/10.1016/j.conbuildmat.2018.03.068
0950-0618/

sludge residue but also realizes the stable and highly efficient immobilization of heavy metals in the sludge residue, and thus achieves the safe treatment of sludge residue [4–8]. In these new trials, using sludge residue to prepare geopolymer has become a new choice.

Geopolymer is regarded as an inorganic polymer gelled material; its most promising development and application prospects are for use with inorganic nonmetallic materials [9,10]. The sludge residue has the same components and structures as other raw materials that are used to synthetize geopolymer, and therefore, it can be used to prepare geopolymer alone or with other raw materials [11,12], and thus it may be able to realize the safe reduction and reclamation of sludge residue. In the meantime, the peculiar cage structure and microcrystalline structure in the prepared geopolymer prepared may well immobilize the heavy metals in the sludge residue [13], which reduces the leaching risk of heavy metals, achieving the hazard-free treatment of sludge residue. Therefore, we concluded that geopolymer preparation a reasonable choice for the disposal of sludge residue. Thus far, no studies on preparing geopolymer with sludge residue have been reported. Most related studies use a mixture of sludge incineration residue and alkali-active materials (such as fly ash, slag, and kaoline) to prepare geopolymer [14–16].

Sludge residue has certain amounts of silicon dioxide (SiO_2) and iron (III) oxide (Fe_2O_3) crystals, and the existence of these crystals destroys the structure of geopolymer during the process of preparing the polymer, influences the dissolution of silicon (Si) and aluminum (Al) elements and hinders thorough inorganic polymerization reactions. After the sludge residue is pretreated by means of incineration, the contents of SiO_2 and Fe_2O_3 crystals in the residue are reduced significantly, which enables the Si and Al elements to exist in the sludge residue with an amorphous form and makes it easy to dissolve out, thereby enhancing the physicochemical properties of the sludge residue in the process of geopolymer preparation and increasing the synthetic speed and compressive strength of geopolymer [17]. The air pollution problem caused by the dioxin produced during the incineration of sludge, however, cannot be ignored, as it restricts the application of sludge incineration residue to the preparation of geopolymer. Therefore, if the sludge residue can be pretreated by other means before the preparation of geopolymer to reduce the contents of crystalline structures, the air pollution problems caused by the incineration can be avoided and will provide technical support for the use of sludge residue to prepare geopolymer. Studies on the use of other means to pretreat sludge residue to prepare geopolymer have not yet been reported.

Currently, methods that can be used to pretreat such solid waste as fly ash, coal ash, cinder, slag, and domestic garbage include physical and chemical methods [18]. Among these methods, the physical pretreatment method mainly refers to a method that uses ball milling under shear force to incur the dislocation and transformation in the internal of crystals contained in the sludge residue, to destroy the structural compositions of the crystals, enlarge the specific surface area, and increase the physicochemical properties during the synthetic reaction. The chemical pretreatment method refers to the method that mixes the solid residue particles with acid (hydrochloric acid or sulfuric acid), alkali (lime-stone or solid particle), or sulfate to strengthen the dissolution of Si and Al in the residue and to increase the physicochemical properties during in the synthetic reaction. As seen from existing research reports, in studies on the pretreatment of fly ash, domestic garbage, cinder, and zeolite, the wet alkalizing method (using the NaOH solution) has certain advantages over other pretreatment methods (such as the dry alkalizing method and ball milling) in terms of particle crushing, activation of components, dissolution of heavy metals, and treatment fees [19,20]. The wet alkalizing method, as a better choice for the pretreatment of sludge residue, can be used to prepare geopolymer.

This work used the wet alkalizing method to pretreat sludge residue and took slag as the gelled material to prepare geopolymer. First, we designed orthogonal experiments to study the effects of the sludge residue/NaOH ratio, calcination temperatures, and heat preservation time on the pretreatment of sludge residue to obtain the best pretreatment parameters of the sludge residue. Then, we studied the influences of different dosages of the alkali activator, the silicate modulus of the alkali activator, and the doping amount of the pretreated sludge residue on the compressive strength of the synthetized geopolymer, and we determined the best combination of parameters to prepare the sludge residue–based geopolymer. Last, based on the analysis of differences in the species distribution of heavy metals in the sludge residue, we explored the immobilization mechanisms and efficiencies of heavy metals in the geopolymer, which provided a technical support and theoretical basis for the reclamation of sludge residue.

2. Materials and methods

2.1. Materials

The sludge residue used in this experiment was obtained from a wastewater treatment plant in Shenzhen. The slag used as the gelled material in this study was the granulated slag from the quick cooling of a smelter cinder during the process of manufacturing steel. Laboratory-grade NaOH and sodium silicate (containing 8.5 wt% Na_2O, 27.3 wt% SiO_2, and 64.2 wt% H_2O) were used as the alkaline activators. The basic properties of sludge residue and slag, including particle size distribution, the metallic oxides, and the heavy metals, were analyzed by particulate size description analyzer, X-ray diffraction (XRD), and X-ray fluorescence (XRF), with results as shown in Table 1. As seen, the sludge residue and slag contained a certain amount of SiO_2 and aluminum oxide (Al_2O_3), indicating that the chemical components of sludge residue and slag used in this work were similar to those of general gelled materials. Additionally, the particle sizes of the sludge residue and slag were as small as 1.13–16.13 μm and 3.69–39.63 μm, which were used to prepare the geopolymer. The observation results from the polarizing microscope indicated that different light refraction effects occurred in the sludge residue and plenty of spherical particles were contained therein, whereas no obvious spherical particles and different light refraction effects were in the slag. This finding indicated that the sludge residue contained plenty of crystals (such as SiO_2), whereas few or no crystals existed in the slag. Therefore, the pretreatment of sludge residue was required before being used to prepare the geopolymer.

Table 1
Properties of sludge residue and slag.

Materials		Sludge residue	Slag
Particle size distribution	Mean diameter (μm)	13.96	16.39
	Median diameter d50 (μm)	3.39	13.50
	Butt diameter d96 (μm)	16.13	39.63
Chemical components (wt.%)	SiO_2	33.61	38.50
	Al_2O_3	39.39	13.40
	CaO	13.63	6.30
	Fe_2O_3	3.96	23.50
	SO_3	1.30	4.72
	P_2O_5	1.39	10.10
	MgO	3.63	1.81
	K_2O	2.03	0.99
	Others	1.16	0.68
Heavy metals (mg/kg)	Cr	89.34	1.39
	Cd	393.36	6.39
	Cu	1077.52	3.96
	Zn	2726.11	1.68
	Ni	92.85	1.94
	Pb	7059.22	13.62