ARTICLE IN PRESS

Waste Management xxx (2018) xxx-xxx



Contents lists available at ScienceDirect

Waste Management



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

Formulation of sludge incineration residue based geopolymer and stabilization performance on potential toxic elements

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ARTICLE INFO

Article history: Received 11 October 2017 Revised 10 April 2018 Accepted 16 April 2018 Available online xxxx

Keywords: SIR-based geopolymer Formulation Compressive strength Potential toxic elements Stabilization

ABSTRACT

Using sludge incineration residue (SIR) to prepare SIR-based geopolymer can realize the minimization and reclamation of urban sludge as well as the stabilization of potential toxic elements. This work has researched the critical factors that influence the formulation of SIR-based geopolymer and the stabilization performance on potential toxic elements. Results showed that the addition amount of SIR had significant influence on the compressive strength of geopolymer, while the modules of sodium silicate and the sodium silicate/metakaolin ratio had little influence thereon. When the addition amount of SIR was 10%, the modules of sodium silicate was 1.3 and the sodium silicate/metakaolin ratio was 0.86, the compressive strength of geopolymer reached the maximum of 95.6 MPa. When the temperature increased from 25 °C to 800 °C, the compressive strength showed a tendency of increasing first and then decreasing. SIRbased geopolymer could significantly reduce the leaching of Zn and Cu from 1264.8 mg/kg and 856.3 mg/ kg to 50.8 mg/kg and 30.7 mg/kg respectively, leading to a stabilizing efficiency more than 96%. pH influenced the stabilization of geopolymer on the potential toxic elements, the effective diffusion coefficients *De* of Zn, Cu, Ni, Pb and Cr at pH = 4 were higher than those at the pH of 7, which were probably due to the physic-chemical characteristics and the corresponding changes in the leaching and species distribution of the potential toxic elements.

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1. Introduction

Residual sludge, as an inevitable product during sewage treatment, brings serious environmental pollution problems. How to scientifically treat the residual sludge with large output, complex ingredients and rich potential toxic elements, in order to realize the minimization, harmlessness and reclamation, has become one of the most important subjects in the environmental field (Breulmann et al., 2015; Peccia and Westerhoff, 2015; Trung et al., 2013). Sludge disposal alternatives include landfill,

https://doi.org/10.1016/j.wasman.2018.04.022 0956-053X/© 2018 Elsevier Ltd. All rights reserved. agricultural application and incineration (Dai et al., 2014; Fonts et al., 2008). Although landfill and agricultural application are considered as the more economical and commonly methods, they still have such disadvantages as land occupation and soil pollution (Sánchez-Monedero et al., 2004). To date, incineration has become one of the most widely used technologies in residual sludge disposal. On one hand, such technology could significantly reduce the volume of residual sludge produced from sewage treatment plants, on the other hand, the reclamation of sludge incineration residue (SIR) also has huge economic and environment-friendly benefits.

However, as is well known, the SIR is rich in the potential toxic elements such as Zn, Cu, Cr, Cd, Ni, Pb, and hence the potential toxic elements contained therein must be stabilized prior to the reclamation of SIR to make it reach the reclamation standard. Cement-based stabilization technology is one of the most widely used methods for the stabilization and reclamation of the

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hazardous waste with potential toxic elements (Voglar and Leštan, 2010). However, this technology requires high temperature (as high as 1400 °C) during cement production, which contributes large emissions of greenhouse gases. In the meantime, the cement with SIR has weak mechanical property, low acid and alkali resistance, high temperature resistance and anti-carbonation, and high leaching risks of potential toxic elements, which is easy to cause second pollution and hard to be used (Cyr et al., 2007; Naamane et al., 2016; Song et al., 2013). As a result, people gradually try to prepare gel materials through other means, namely mixing the sludge incineration residue with granular alkali-activated substances (including fly ash, blast furnace residue, metakaolin, kaolinite clay and red mud), to prepare geopolymer (Chen et al., 2011; Santa et al., 2013; van Jaarsveld et al., 2002; Wang et al., 2005; Yan and Sagoe-Crentsil, 2012), in order to realize the minimization and reclamation of sludge.

In recent years, the method that uses SIR to prepare geopolymer for sludge reclamation has been proved to have high research and practical values in the aspect of residual sludge minimization. The related research results show that different gel materials for preparing geopolymer render the prepared geopolymer to have obvious structure differences (Zhang et al., 2017). Compared with other gel materials, alkali-activated gelled material has good durability, acid and alkali resistance and could prepare stable geopolymer by mixing with SIR, and in addition, it may increase the mechanical property and stability of geopolymer in a certain extent by optimizing the process parameters of formulation (Yan and Sagoe-Crentsil, 2012). However, there still exists the environmental risk caused by the leaching of potential toxic elements in SIR-based geopolymer. The existing researches on the stabilization performance of geopolymer on potential toxic elements basically focus on the geopolymer prepared from kaolin, metakaolin or fly ash, and as seen from the research results, the Calcium-Silicate-Hydrate (C-S-H) gel in the geopolymer that has strong gelling property and stability can wrap the SIR particles and thereby realize the stabilization of potential toxic elements (Zhang et al., 2014). However, the chemical decomposition of the gel significantly reduces the stabilization performance on potential toxic elements. In addition, researches also point out that different kinds and contents of residual materials for formulation have significant effects on the stabilization performance on potential toxic elements of the prepared geopolymer (Malvifya and Chaudhary, 2006; Wang et al., 2005), and what's more, the parameters such as the concentrations of potential toxic elements and the pressure control in the formulation process also have significant effects on the stabilization performance (Al-Harahsheh et al., 2015; Cheng et al., 2012). Till now, although Chen et al. (2011) and Santa et al. (2013) have studied the characteristics and stabilities of different kinds of sludge-based geopolymer, the characteristics and the functional mechanisms of the potential toxic elements stabilization in the sludge-based geopolymer are still not clear.

This study chose the alkali-activated metakaolin as gel material to formulate SIR-based geopolymer, the change rules of mechanical properties under different addition amounts of SIR, modules of sodium silicate and sodium silicate/metakaolin ratios were studied by means of orthogonal test, in order to determine the optimal parameter portfolio for the formulation of SIR-based geopolymer. The stability of geopolymer was also observed by analyzing the changes of the compressive properties thereof at different temperatures. Based on the analysis of specification distribution of the potential toxic elements in SIR and the study on the stabilization performance of the geopolymer on potential toxic elements such as Zn, Cu, Cr, Cd, Ni, Pb under different temperatures and pH values, this study also analyzed the stabilizing mechanism and efficiency of geopolymer on the potential toxic elements in SIR, so as to provide technological support and theoretical basis for the minimization, harmlessness and reclamation of residual sludge.

2. Materials and methods

2.1. Materials

The sewage sludge, obtained from a sewage treatment plant in Shanghai, China, was calcined at the temperature of 800 °C for 1 h to produce a proper chemical composition for synthesizing geopolymer. Metakaolin was used as the main gel material. Laboratory-grade NaOH and sodium silicate (8.5 wt% Na₂O, 27.3 wt% SiO₂, and 64.2 wt% H₂O) were used as alkaline activators. The basic properties of the metakaolin and SIR were analyzed by particulate size description analyzer (S3500, Microtrac, USA), Xray fluorescence (XRF, S4 Explorer, Bruker, Germany) and X-ray diffraction (XRD, DX-2500, Rigaku, Japan), including the particle size distribution, the potential toxic elements contained in the SIR and the metakaolin, as shown in Table 1. As seen, the SIR rich in SiO₂ and Al₂O₃ had chemical components similar to those of ordinary gel materials, and could be used for preparing geopolymer due to its alkali-activated activity. The concentrations of the potential toxic elements in SIR differed a lot, among which, the concentration of Zn reached 3334.1 mg/kg, which rendered large environmental pollution risk, besides that, certain amount of Cu, Cr. Ni. Pb and Cd also existed therein.

2.2. Experimental designs

In this work, the influences of the modules of sodium silicate, the addition amount of SIR and the sodium silicate/metakaolin ratio, which were signed as A, B and C respectively, on the mechanical properties of the SIR-based geopolymer were studied. The SIR and the metakaolin were weighted and mixed with the NaOH and sodium silicate using a mechanical stirrer for 2 min in slow speed and 3 min in high speed until well combined. Then the mixture was poured into steel containers ($30 \times 30 \times 30$ mm) and vibrated for 50 times by using a vibration table to remove entrapped air bubbles. The geopolymer samples were cured for 24 h at an ambient temperature of 25 °C, humidity of 100%. The change ranges of each parameter were shown in Table 2.

Table 1
Properties of sludge incineration residue and metakaolin.

Materials		SIR	Metakaolin
Particle size distribution	Mean diameter (µm) Median diameter d50 (µm)	26.9 23.2	7.9 7.5
	Butt diameter d97 (µm)	74.0	16.2
Chemical components (wt%)	SiO ₂	38.5	50.6
	Al_2O_3	13.4	46.8
	CaO	6.3	0.6
	Fe ₂ O ₃	23.5	0.1
	SO ₃	4.7	0.5
	P_2O_5	10.1	0.6
	MgO	1.8	0.1
	K ₂ O	1.0	0.2
	Others	0.7	0.5
Potential toxic elements (mg/kg)	Cr	489.3	-
	Cd	20.8	-
	Cu	976.5	-
	Zn	3334.1	-
	Ni	268.0	-
	Pb	103.4	-

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