Process Safety and Environmental Protection 117 (2018) 684–693

Contents lists available at ScienceDirect

Process Safety and Environmental Protection

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

# Potential solidification/stabilization of clay-waste using green geopolymer remediation technologies

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ARTICLE INFO

*Article history:*
Received 13 October 2017
Received in revised form 13 June 2018
Accepted 13 June 2018
Available online 21 June 2018

*Keywords:*
Solidification/stabilization
Green remediation
Clay-waste
Mechanical properties
Si/Al ratio
Leaching
Heavy metals
SEM-EDX

ABSTRACT

Solidification and stabilization is a remediation technique for hazardous wastes that promotes resource recycling and reduces the environmental burdens of waste management. To accomplish this successfully, different types of wastes or hazardous materials are treated with different binders and techniques. This study proposed a green remediation approach to treat and recycle clay-waste by using solidification/stabilisation which is enhanced by the injection of two chemical solutions, sodium hydroxide (NaOH) and sodium silicate ($Na_2SiO_3$) and partial addition of low-calcium class F fly ash. The unconfined and compressive strength tests were conducted to investigate the mechanical properties of the developed geopolymer pastes at different time conditions. The change in the microstructures was characterized by using Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Microanalysis (EDX). The effects of Si/Al ratio on the compressive strength and the microstructure of the solidified geopolymer product are also summarized in this paper. In addition, the normalized weight variations after 28 days curing in deionized water (pH = 7.0) was highlighted and the leaching behavior of heavy metals in the clay-waste based geopolymer specimens after soaking up to 14 days was examined with atomic absorption spectroscopy. Experimental results demonstrate the potential treatment, solidification and stabilization of the clay-waste by employing geopolymer remediation technologies and the optimum compressive strength can be obtained with a ratio of 30% fly ash. The collected clay-waste has strength of up to 5–20 MPa at 28 days curing which meet suitable mechanical properties and compact microstructure characterization.


## 1. Introduction

During the past decade, the concept of sustainable development has continued to improve and this has allowed geopolymerization technology to emerge as a possible solution for the effective solidification and stabilization of toxic and waste materials. The principle of solidification/stabilization (S/S) aim to immobilize the toxic constituents of hazardous wastes to prevent their leaching from the wastes once disposed. It involves both physical and chemical process formulations that is accomplished by reducing the solubility of the waste components, and by physically isolating the waste and decreasing its surface area. The material used for S/S not only stabilizes the hazardous waste by chemical means but also insolubilizes, immobilizes by solidifying them, encapsulates, destroys, sorbs, or otherwise interacts with the selected waste components (Rachana and Rubina, 2006).

The solidification/stabilization (S/S) technology is most suitable for treating inorganic wastes and has received a lot of attention in recent years. Researchers have investigated a range of wastes for which proportions of cement wastes, and fillers are used in solidification/stabilization and they have consistently produced satisfactory results (Peng et al., 2015; Viguri et al., 2001; Du et al., 2014; Zhang et al., 2010a; Naamane et al., 2016; Li et al., 2015; Elzbieta et al., 2014). On the other hand, the cement industry have been found to be a highly energy intensive industry acting as a major source for carbon dioxide emission that leads to some serious environmental hazards such as global warming and large amounts of carbon dioxide ($CO_2$) production (Imbabi et al., 2012). Extensive research works were carried out to demonstrate the performance of industrial waste byproduct materials used as reactive binders for S/S of contaminated soil with good strength, durability and economic properties (Lei et al., 2015; Mijno et al., 2007; Ogundiran et al., 2013; Arun and Sivashanmugam, 2015; Shi and Fernandez-Jimenez, 2006; Zhang et al., 2010b; Li and Poon, 2017).

Several materials containing large amounts of silica and alumina that partially dissolve in alkaline solutions have been used as reagents for geopolymerization reactions. These include blast

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https://doi.org/10.1016/j.psep.2018.06.013
0957-5820/

furnace slag, coal fly ash, phosphorus slag, steel slag, metakaolin, CDG, loess, and so one. Also, a combination of two or more of them can be used as an alternative to Portland cement (Li and Poon, 2017; Dassekpo et al., 2017a; Dassekpo et al., 2017b). Although geopolymerization is not a new concept, the application of this technology to remediate various waste materials is relatively little. Thus, only in the last decade has it been applied to fly ash. A typical fly ash-based geopolymer mix consists of approximately 60% mass of dry fly ash, and approximately 15% mass of dry additional Al–Si source materials, and the rest of the mix is the alkali activators solution (van Jaarsveld et al., 1998). However, this percentage can vary greatly depending on the characteristics needed and that includes the Si and Al contents of the fly ash. Thus, percentages of fly ash as low as 10% have been employed in some geopolymer reactions (Bankowski et al., 2004a; Bankowski et al., 2004b).

In addition, the contamination risk of geopolymer to surrounding soils and its suitability in immobilizing toxic metals and the leaching behavior of synthesized geopolymers from different raw materials was investigated (Aly et al., 2008; Temuujin et al., 2011; Izquierdo et al., 2009; Alvarez-Ayuso et al., 2008; Arioz et al., 2012; Fernando and Said, 2010). Álvarez-Ayuso et al. (Alvarez-Ayuso et al., 2008) studied the leaching behavior of fly ash based geopolymer in deionized water, and found the leaching ability of metals depended on the type of chemical elements. Aly et al. (Aly et al., 2008) examined the leaching ability of Cs, Sr, Na and Al from metakaolin based geopolymer powder in deionized water, where a small amount of Cs and Sr were added in the geopolymers. They found that Cs and Sr were well and partly bound with geopolymers. Izquierdo et al. (Izquierdo et al., 2009) have investigated the leaching tests on fly ash-slag based geopolymer in deionized water and found that geopolymers can provide an efficient encapsulation for heavy metals once the dosage, synthesis and curing conditions were well tailored for the synthesis of geopolymers.

In light of all the above, a systematic study was conducted to treat and recycle clay-waste for its potential utilization by using green geopolymer remediation stabilization/solidification approaches. The Unconfined Compressive Strength (UCS) and the standard compressive strength test were carried out to study the mechanical performance of the developed S/S products. The Scanning Electron Microscopy (SEM) and EDX microanalysis imaging were also applied on the developed material to characterize their microstructure and the effect of Si/Al ratio on their strength performance. In addition, the leaching behavior of the synthesized geopolymer pastes was also investigated to access the contamination risk.

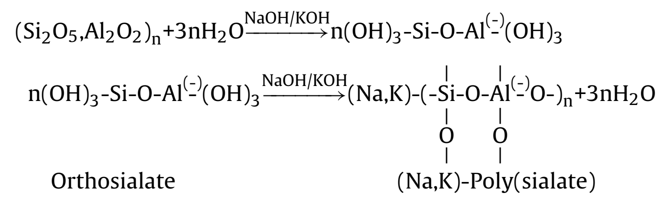
## 2. Geopolymerization reaction and formulation

The geopolymerization reaction is essentially based on the synthetic alkali aluminosilicate material produced from the chemical reaction of a solid aluminosilicate with a highly concentrated alkaline hydroxide and/or silicate solution. This reaction can be achieved at either ambient or elevated temperatures (Hansen, 1992). The geopolymerization involves the dissolution of silica and alumina compounds in the alkaline solution and then bonding to each other to form a polymeric structure. According to the research work of Duxson et al. (Tavakoli and Soroushian, 1996) and Dimas et al. (Sagoe-Crentsil et al., 2001), the geopolymerization process includes the dissolution of solid aluminosilicate materials into a strong concentration of alkaline solution, the formation of silica-alumina oligomers, the polycondensation of oligomeric species to form inorganic polymeric material, and the bonding of undissolved solid particles in the final geopolymeric structure. The general geopolymers chemical formula can be written as follow:

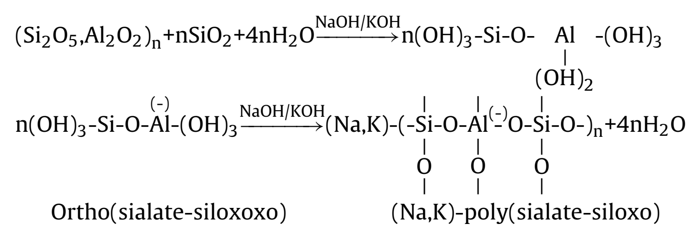
$$M_n[-(Si-O_2-)_z-Al-O_2-]_n \bullet wH_2O \quad (1)$$

where $M$ is the cation (sodium or potassium), $n$ is the degree of polymerization, $z$ is the quantifying factor for amount of $SiO_2$ monomer units (typically 1, 2, or 3), and $w$ is the amount of binding water, which can be up to 7 (Shayan and Xu, 2003). Based on $z$ value, three types of oligomers can be formed: poly sialate (PS) (-Si-O-Al-O-), poly sialate-siloxo (PSS) (Si-O-Al-O-Si-O), and poly sialate-disiloxo (PSDS) (Si-O-Al-O-Si-O-Si-O) which is obtained by packing $SiO_2$ in the polymeric network of PSS and has the highest density and the lowest porosity (Tam et al., 2005). Each type of the oligomers has certain Si/Al ratio that means that, the type of the oligomer can be identified by the ratio of constituting silica and alumina species.

According to Joseph Davidovits, the hardening mechanism for geopolymerization is based essentially on the polycondensation reaction of the geopolymeric precursors, usually alumino-silicate oxides with alkali polysilicates yielding polymeric Si-O-Al bonds (Davidovits, 1994) as is described below:



(2)



(3)

## 3. Materials and methodology

### 3.1. Materials

The clay used in this study was collected from Shenzhen Bay construction site. The low-calcium class F fly ash conforming to ASTM C-618 specification (ASTM C618-08a, 2008) supplied from Shenzhen coal power plants in China served as a complement precursor in the solidification/stabilization process (Fig. 1). The geopolymerization reaction was produced by mixing clay-waste and clay-waste-fly ash with sodium hydroxide (NaOH) and sodium silicate ($Na_2SiO_3$). The solutions were prepared by dissolving sodium hydroxide in pellets form with 98% purity in water to form 14 M molarity concentration and mixed with sodium silicate solution.

### 3.2. Mix design

The mix design adopts in this research is presented in Table 1. The main materials used are clay-waste with partial addition of fly ash ratio of 0%, 10%, 20% and 30%.

### 3.3. Solidified specimens preparation

The solidified specimens were prepared as paste using plastic molds of 70 mm cubes by mixing until homogenous and compacted using a vibrating table and hand tamping as and when needed. Three replicate samples i.e. 9 cubes for each mixture ratio were