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Durability and leaching behavior of mine tailings-based geopolymer bricks

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HIGHLIGHTS

- Systematically studied durability and leaching behavior of MT-based geopolymer bricks.
- Immersion in pH = 4 and 7 solutions leads to substantial strength loss.
- Immersion in pH = 4 and 7 solutions leads to minor water absorption and weight loss.
- MT-based geopolymer bricks effectively immobilize the heavy metals in MT.
- FRDM can satisfactorily describe the leaching behavior of heavy metals.

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ABSTRACT

Disposal of mine tailings (MT) in impoundments may have adverse environmental impacts such as air pollution from dust emissions and release of heavy metals to surface and underground water. Geopolymerization as an environmentally-friendly and sustainable method has been used to stabilize MT so that they can be used as construction material. In this paper, the durability and leaching behavior of MT-based geopolymer bricks are studied by measuring unconfined compression strength (UCS), water absorption, weight loss, and concentration of heavy metals after immersion in pH = 4 and 7 solutions for different periods of time. Microscopic/spectroscopic techniques, SEM, XRD and FTIR, are also employed to investigate the change in microstructure and phase composition of MT-based geopolymer bricks after immersion in the solutions. To describe the leaching behavior of MT-based geopolymer bricks, the first order reaction/diffusion model (FRDM) is used to analyze the leaching test data. The results indicate that although there is a substantial strength loss after immersion in pH = 4 and 7 solutions, the water absorption and weight loss are small. The strength loss is mainly due to the dissolution of geopolymer gels as indicated by the microscopic/spectroscopic analysis results. The leaching analyses show that the heavy metals are effectively immobilized in the MT-based geopolymer bricks, which is attributed to the incorporation of heavy metals in the geopolymer network. The FRDM can satisfactorily describe the leaching behavior of heavy metals in the MT-based geopolymer bricks and the analysis results indicate that the solubility or reaction rate is an important factor controlling the leaching behavior.

1. Introduction

Mine tailings (MT) are a major waste material generated by mining operations. In current practice, MT are transported in slurry form to and deposited in storage impoundments. Storage of MT in such impoundments leads to occupation of large areas of land, costly construction and maintenance, and potential environmental and ecological risks. MT can cause air pollution due to dust emissions resulted from surface erosion. MT can also pollute surface and underground water due to the leaching of heavy metals. The sulfide minerals in MT such as pyrite (FeS_2), pyrrhotite ($Fe_{1-x}S$), and chalcocite (Cu_2S) oxidize in the presence of air and water, yielding sulphuric acid and releasing metallic oxides such as FeO [1]. This phenomenon, known as acid mine drainage (AMD), leads to the drop of pH and results in further leaching of heavy metals such as Cd, As and Cu [2]. AMD has caused serious contamination of surface and underground waters in the United States [3]. Therefore, it is vital to take measures to reduce the risk of environmental contamination by MT.

Generally, there are three methods to reduce the potential environmental hazards imposed by MT: (1) isolation of MT, (2) chemical stabilization of MT, and (3) a combination of these two methods [2,4]. The isolation techniques include containment of MT from the surrounding environment such as capping the tailings impoundment surface. This can be achieved by designing and

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constructing a closure system similar to that used for landfills [5]. For chemical stabilization, chemicals or cementitious materials are added to immobilize the heavy metals in MT through physical encapsulation and/or chemical reactions. In current practice, pozzolanic materials such as cement and lime are commonly used to stabilize MT [1,6–11] although other materials such as fly ash, slag and aluminum are also studied by researchers [12,13]. The isolation and stabilization techniques can also be used simultaneously. For example, the tailings surface can be treated by binders such as organic polymers, water glass and Portland cement to improve surface erosion resistance and reduce water infiltration, and, in the meantime, the hardened surface acts as a capping system which isolates the underlying tailings from the surrounding environment [14].

Since the stabilization of MT based on the reaction with calcium has a number of limitations, such as inferior mechanical properties, low acid resistance, poor immobilization of contaminants at high concentrations, and more importantly high-energy usage and greenhouse gas emissions related to production of Portland cement [15,16], researchers have studied other stabilization methods to stabilize MT [1,13,17,18]. Of these different methods, geopolymerization is a promising one to effectively stabilize MT in an economical and environmentally-friendly way. In this method, geopolymer gels are produced on the MT particle surface and the newly formed gels bind the particles together. Geopolymer is an amorphous binder with a polymeric network structure consisting of repeating units of —Si—O—Al—O— and is formed by alkali activation of silica and alumina containing materials at high pH and room or slightly elevated temperature [19]. Geopolymer binder offers superior mechanical properties, excellent durability, and effective immobilization of heavy metals [19–28]. An extensive research has been conducted on fly ash-, slag-, and metakaolin-based geopolymers [21,24,28–32] and MT-based geopolymer has recently attracted attention of researchers worldwide [18,22,33–38]. Van Jaarsveld et al. [18] studied the feasibility of using MT-based geopolymer paste as a cover system for tailings dam. The results indicated that fly ash and as much as 65–70% MT can be used to produce geopolymeric material suitable for capping mine tailings. Pacheco-Torgal et al. [22] evaluated the durability and environmental performance of calcined tungsten MT-based geopolymer (CTMTG) and reported that the CTMTG binder exhibits better durability than Portland cement binder and the concentrations of released heavy metals are all below the DIN limits. Giannopoulou and Panias [33] showed that the compressive strength of mixed fly ash and MT-based geopolymer increases with the fly ash content and the concentrations of leached heavy metals in neutral and acidic solutions are all below the Greek Standard limits. Silva et al. [34] studied the stability of CTMTG immersed in water and reported disintegration after a certain period of time mainly due to deficient geopolymeric reaction.

Ahmari and Zhang [39] studied the production of eco-friendly bricks based on geopolymerization of copper MT, focusing on their physical and mechanical properties. The results indicate that by properly selecting the preparation condition (initial water content, NaOH concentration, forming pressure and curing temperature), MT-based geopolymer bricks can be produced to meet the ASTM requirements on physical and mechanical properties. In this paper, the durability and leaching behavior of the copper MT-based geopolymer bricks are studied.

2. Experimental

2.1. Materials

The materials used in this investigation include copper mine tailings (MT), reagent grade 98% sodium hydroxide (NaOH), de-ionized water, and BDH Aristar Plus (67–70%) nitric acid. The MT were received as dry powder from a local mine company in Tucson, Arizona. Table 1 shows the chemical composition of the MT. It can be seen that the MT consist of mainly silica and alumina with substantial amount of calcium and iron. Grain size distribution analysis was performed on the MT using mechanical sieving and hydrometer analysis following ASTM D6913 and ASTM D422. Fig. 1 shows the particle size distribution curve. The mean particle size is around 120 μm with 36% particles passing No. 200 (75 μm) sieve. The specific gravity of the MT particles is 2.83.

The sodium hydroxide (NaOH) flakes were obtained from Alfa Aesar Company in Ward Hill, Massachusetts. The sodium hydroxide solution is prepared by dissolving the sodium hydroxide flakes in de-ionized water. The nitric acid (HNO_3) was manufactured by BDH and supplied by VWR.

2.2. Preparation of brick samples

First, the MT were mixed with NaOH solution. The NaOH solution was prepared by adding NaOH flakes to de-ionized water and stirring for at least five minutes. Due to the generated heat, enough time was allowed for the solution to cool down to room temperature before it was used. The NaOH solution was slowly added to the dry MT and mixed for 10 min to ensure the homogeneity of the mixture. The generated mixture exhibited varying consistency depending on the initial water content. The mixture's consistency varied from semi-dry to semi-paste as the water content increased from 8% to 18%. The mixture was then placed in Harvard miniature compaction cylindrical molds of 33.4 mm diameter and 72.5 mm height with minor compaction. The compacted specimens were compressed with an ELE Tri Flex 2 loading machine at different loading rates to ensure that the duration to reach the specified forming pressure was about 10 min for all the specimens. After compression, the specimens were de-molded and placed uncovered in an oven for curing until tested.

Table 1
Chemical composition (wt.%) of mine tailings.

Chemical compound	Content[a] (%)	Standard deviation (%)
SiO_2	64.8	2.08
Al_2O_3	7.08	0.70
Fe_2O_3	4.33	0.71
CaO	7.52	1.06
MgO	4.06	0.93
SO_3	1.66	0.31
Na_2O	0.90	0.23
K_2O	3.26	0.42
Trace elements		
Pb	0.000286	0.0007
Zr	0.012	0.001
Mo	0.022	0.003
Zn	0.068	0.009
Cu	0.076	0.009
Mn	0.163	0.034
Ti	0.213	0.006

[a] The values are the average of seven tailings samples.

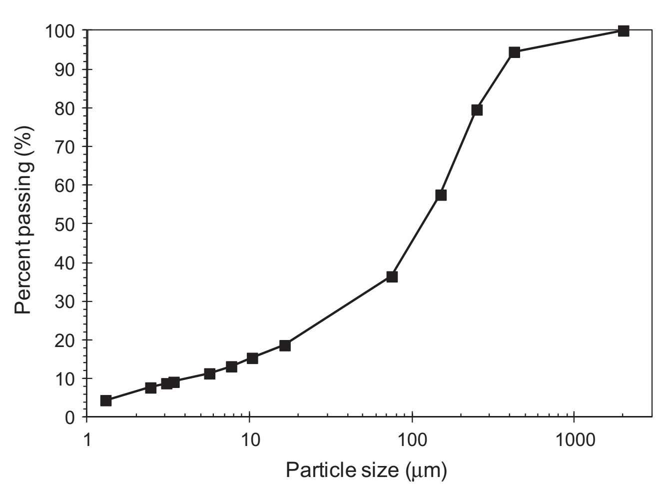

Fig. 1. Particle size distribution of MT powder.