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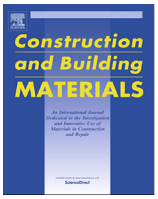

Leaching and microstructural properties of lead contaminated kaolin stabilized by GGBS-MgO in semi-dynamic leaching tests

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HIGHLIGHTS

- Leaching is diffusion-controlled at pH 7 and 4 while dissolution-controlled at pH 2.
- Observed diffusion coefficient is affected by the SAR pH and binder dosage.
- Pb is primarily precipitated as hydrocerussite ($Pb_2(CO_3)_2(OH)_2$) in soil matrix.
- ANC and pore structure of the treated soil affect diffusive properties of Pb.

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ABSTRACT

Ground granulated blast furnace slag (GGBS) is widely used to stabilize soils due to its environmental and economic merits. The strength and durability of reactive MgO activated GGBS (GGBS-MgO) stabilized lead (Pb)-contaminated soils have been explored by previous studies. However, the effects of simulated acid rain (SAR) on the leachability and micro-properties of GGBS-MgO stabilized Pb-contaminated soils are hardly investigated. This research studies the leachability and microstructural properties of GGBS-MgO stabilized Pb-contaminated kaolin clay exposed to SAR with initial pH values of 2.0, 4.0 and 7.0. A series of tests are performed including the semi-dynamic leaching tests using SAR as the extraction liquid, acid neutralization capacity (ANC), mercury intrusion porosimetry (MIP), and X-ray diffraction (XRD) tests. The results demonstrate that as the SAR pH decreases from 7.0 to 4.0, the Pb cumulative fraction leached (*CFL*) and observed diffusion coefficient (D^{obs}) increases significantly whereas the leachate pH decreases. Meanwhile, increasing the GGBS-MgO content from 12% to 18% results in the decrease of *CFL* and D^{obs}. Further decreasing the SAR pH to 2.0 results in the dissolution-controlled leaching mechanism regardless of the binder dosage. The differences in the leaching properties under different pH conditions are interpreted based on the cemented soil acid buffering capacity, hydration products and pore size distributions obtained from the ANC, MIP, and XRD tests, respectively.

1. Introduction

Numerous abandoned industrial sites worldwide have been found to be contaminated with a wide range of heavy metals [1–7]. These toxic metals such as lead (Pb), zinc (Zn), copper (Cu), and cadmium (Cd), if treated improperly, can pose severe threats to the environment and human health. Considering the fast urbanization and ever-increasing value of the land resources, particularly in the developing countries such as China and India, it is imperative to develop effective and economical technologies to remediate these heavy metal contaminated industrial sites. The ultimate goal is to eliminate their negative environmental impact to the society and improve the mechanical properties of soils to facilitate post-construction. Solidification/Stabilization (S/S) has been widely used to immobilize contaminants and improve the soil properties [2–3,8]. After S/S, the remediated soils can be reused in-situ as engineering construction materials, which would help on the fast redevelopment of the contaminated site [9–10].

Portland cement (PC) is the most popular binder used in S/S [11]. However, its manufacturing process is associated with high power consumption (5000 MJ/t PC), non-renewable resources usage (1.5 t limestone and clay/t PC) and considerable emissions

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of carbon dioxide (CO_2), dust, and deleterious gases (SO_2, CO, NO_X) (0.95 t/t PC) [12–13]. Therefore, full or partial replacement of PC by more sustainable industrial by-products (e.g. fly ash and slag) as alternative binders in treating contaminated soil has received ever-increasing popularity. One of the promising alternative binders is alkali-activated slag (AAS) cement using ground granulated blast furnace slag (GGBS) as the main raw material. However, several drawbacks are associated with the utilization of AAS in S/S including over-rapid setting, difficulty in handing/transporting the caustic alkalis and uneconomical efficiency [12]. To address these issues, reactive magnesia (MgO) has been used as an effective activator for the GGBS [12,14–16]. Existing studies on the GGBS-MgO binder mainly focus on the strength, durability and microstructural properties of the pastes and stabilized soils [14–16]. The MgO facilities the breakage of Si-O and Al-O bonds in the GGBS to promote the formation of the calcium silicate hydrate (C-S-H) and hydrotalcite ($Mg_6Al_2CO_3(OH)_{16}$)-like phase (Ht) as the main hydration products [17–19] while C-S-H and $Ca(OH)_2$ are the main hydration products in PC stabilized soils [2,21]. The C-S-H and Ht formed would enhance the physical and mechanical properties [14–16,18] and reduce the leachability of contaminants in heavy metal contaminated soils [16,20]. Recently, the feasibility of using this binder for stabilizing heavy metal-contaminated soils has been demonstrated both in the laboratory [20] and a field trial [8]. However, to date, no systematic studies exist on the diffusive properties of heavy metals in GGBS-MgO stabilized heavy metal contaminated soils.

Sharma and Reddy [6] indicate that the acid rain may vary from a highly acidic condition (pH = 2.0) to a neutral condition (pH = 7.0). It is reported that the average pH value of the acid rain in Nanjing City, China is about 5.09 with the lowest pH of 2.89 [21–23]. Du et al. [2] and Yun et al. [24] investigate the leaching behavior and long-term durability of PC solidified/stabilized heavy metal-contaminated soils under various acid rain conditions. They show that heavy metals could be released notably from the stabilized soils with increased acidity. It is expected that due to the different hydration chemistry and reaction products formed in GGBS-MgO and PC binders, the leaching properties of the treated soils exposed to the acid rain would be different. Therefore, it is necessary to comprehensively evaluate the leaching behavior of GGBS-MgO stabilized heavy metal-contaminated soils under different acidic conditions: strongly acidic condition (pH = 2.0), moderate acidic condition (pH = 4.0) and neutral condition (pH = 7.0).

In this study, a series of semi-dynamic leaching tests are performed on lead (Pb)-contaminated kaolin clay using simulated acid rain as the extraction leachant with initial pH values of 2.0, 4.0, and 7.0. The effects of acid rain pH and GGBS-MgO content on the leachability and microstructural properties of the treated soils are studied. The semi-dynamic leaching test results are interpreted by acid neutralization capacity (ANC), mercury intrusion porosimetry (MIP) and X-ray diffraction (XRD). This study provides useful insights for remediating Pb-contaminated clayey soils using the GGBS-MgO binder.

2. Materials and testing methods

2.1. Materials and sample preparations

Kaolin clay is used as a base soil due to its uniform composition (low organic content, homogeneity and uniform mineralogy) and low cation exchange capacity [1–3,14]. The basic physiochemical properties of the kaolin clay are summarized in Table 1. The pH is measured per ASTM D4972 [25] using a pH meter HORIBA D-54. The specific gravity is measured per ASTM D5550 [26]. The Atterberg limits are measured per ASTM D4318 [27]. The kaolin clay is classified as lean clay (CL) based on the Unified Soil Classification System [28]. The moisture content is measured as per ASTM D2216 [29]. The grain size distribution is measured using a laser particle size analyzer Mastersizer 2000.

Table 1
Properties of the kaolin soil used in this study.

Index	Value
pH	8.77
Specific gravity, G_s	2.68
Plastic limit, w_P (%)	14.6
Liquid limit, w_L (%)	29.4
Grain size distribution (%)	
Clay (<0.002 mm)	21.5
Silt (0.002–0.075 mm)	58
Sand (0.075–2 mm)	20.5

Table 2
Main physico-chemical properties of GGBS and MgO.

Property	Value	
	GGBS	MgO
Alkalinity[a]	1.689	–
Reactivity (s)	–	102
Specific surface area (m^2/g)	0.2932	28.023
pH (liquid to solid ratio = 1:1)	10.96	10.53

[a] The alkalinity of the GGBS is defined as the ratio of contents of CaO, MgO, and Al_2O_3 to that of SiO_2.

The physiochemical properties of GGBS and MgO are listed in Table 2. The BET specific surface areas of the GGBS and MgO are measured by nitrogen adsorption using Physisorption Analyzer ASAP2020. The chemical compositions of the kaolin clay, GGBS, and MgO are measured using X-ray fluorescence (XRF) as shown in Table 3. The reactivity of the MgO is measured as the time duration required for the neutralization of an acidic solution (0.25 M acetic acid in this study) by a certain amount of MgO sample (5.0 g in this study) in which phenolphthalein is adopted as the pH indicator [30]. The mean values of the above tests are presented in Tables 1–3.

Pb is used in this study because it is a very common toxic heavy metal in contaminated soils [3,14,31]. Lead nitrate ($Pb(NO_3)_2$) powder (Chemical Analytical Reagent, Sinopharm Chemical Reagent Co., Ltd.) is dissolved in distilled deionized water (DDW) as stock solutions with predetermined Pb concentrations. The simulated acid rain (SAR), used as the extraction liquid (leachant) in the semi-dynamic leaching test, is prepared by diluting nitric acid (HNO_3) and ammonium sulfate ($(NH_4)_2SO_4$) in the DDW. Prior to adding HNO_3, ammonium sulfate ($(NH_4)_2SO_4$) solution is added to the DDW until the concentration of the sulfate ion (SO_4^{2-}) reaches 0.001 mol/L [2]. The stock solutions of SAR are adjusted to three pH value of 2.0, 4.0, and 7.0 respectively. SAR with pH of 2.0 represents a strong acid rain in the field [2].

Previous studies show that a binder with 9:1 ratio of GGBS to MgO (dry weight basis) yields relatively higher strength and lower leachability of stabilized contaminated soils [14]. Therefore, the binder consisting of 90% GGBS and 10% MgO (dry weight basis) is prepared. Three binder contents are set as 12%, 15%, and 18% (dry weight soil basis) which are typical contents in engineering projects [2]. The water content and the Pb concentration are set as 45% and 2% (i.e., 20000 mg/kg) (dry weight soil basis) to simulate a heavily contaminated site soil [11,21], respectively. Six mixtures are investigated in total and denoted as GM_iPb_j, where i = content of the GGBS-MgO binder (i.e., 12, 15 or 18), and j = Pb concentration (%, 0 or 2).

The kaolin clay, GGBS and MgO powders are placed in a plastic bottle and are thoroughly mixed by a bench-top mixer. Then the predetermined volume of Pb $(NO_3)_2$ stock solution is added to the plastic bottle and further mixed for 30 min

Table 3
Chemical compositions of the kaolin soil, GGBS and MgO used in this study measure by XRF.

Chemical composition (*wt*%)	Kaolin	GGBS	MgO
CaO	0.36	33.08	0.84
Al_2O_3	39.3	17.9	0.38
MgO	0.06	6.02	96.5
K_2O	0.21	0.64	0.01
SiO_2	52.1	34.3	1.09
Fe_2O_3	3.38	1.02	0.19
SO_3	0.06	1.64	0.26
MnO	0.11	0.28	0.02
TiO_2	1.12	0.92	0.01
Loss on ignition (at 950 °C)	3.3	4.2	0.7