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1 **Leaching and Microstructural Properties of Lead Contaminated**
2 **Kaolin Stabilized by GGBS-MgO in Semi-Dynamic Leaching Tests**

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

49

50 **Keywords:** Slag; reactive MgO; leaching test; contaminated soil; solidification/stabilization

51 **1 Introduction**

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

65 Portland cement (PC) is the most popular binder used in S/S [11]. However, its
66 manufacturing process is associated with high power consumption (5000 MJ/t PC),
67 non-renewable resources usage (1.5 t limestone and clay/t PC) and considerable
68 emissions of carbon dioxide (CO₂), dust, and deleterious gases (SO₂, CO, NO_x) (0.95
69 t/t PC) [12-13]. Therefore, full or partial replacement of PC by more sustainable
70 industrial by-products (e.g. fly ash and slag) as alternative binders in treating
71 contaminated soil has received ever-increasing popularity. One of the promising
72 alternative binders is alkali-activated slag (AAS) cement using ground granulated

73 blast furnace slag (GGBS) as the main raw material. However, several drawbacks are
74 associated with the utilization of AAS in S/S including over-rapid setting, difficulty in
75 handing/transporting the caustic alkalis and uneconomical efficiency [12]. To address
76 these issues, reactive magnesia (MgO) has been used as an effective activator for the
77 GGBS [12, 14-16]. Existing studies on the GGBS-MgO binder mainly focus on the
78 strength, durability and microstructural properties of the pastes and stabilized soils
79 [14-16]. The MgO facilitates the breakage of Si-O and Al-O bonds in the GGBS to
80 promote the formation of the calcium silicate hydrate (C-S-H) and hydrotalcite
81 ($\text{Mg}_6\text{Al}_2\text{CO}_3(\text{OH})_{16}$)-like phase (Ht) as the main hydration products [17-19] while
82 C-S-H and $\text{Ca}(\text{OH})_2$ are the main hydration products in PC stabilized soils [2,21]. The
83 C-S-H and Ht formed would enhance the physical and mechanical properties [14-16,
84 18] and reduce the leachability of contaminants in heavy metal contaminated soils [16,
85 20]. Recently, the feasibility of using this binder for stabilizing heavy
86 metal-contaminated soils has been demonstrated both in the laboratory [20] and a
87 field trial [8]. However, to date, no systematic studies exist on the diffusive properties
88 of heavy metals in GGBS-MgO stabilized heavy metal contaminated soils.

89 Sharma and Reddy [6] indicated that the acid rain may vary from a highly acidic
90 condition (pH = 2.0) to a neutral condition (pH = 7.0). It is reported that the average
91 pH value of the acid rain in Nanjing City, China is about 5.09 with the lowest pH of
92 2.89 [21-23]. Du et al. [2] and Yun et al. [24] studied the leaching behavior and
93 long-term durability of PC solidified/stabilized heavy metal-contaminated soils under
94 various acid rain conditions. They showed that heavy metals could be released

95 notably from the stabilized soils with increased acidity. It is expected that due to the
96 different hydration chemistry and reaction products formed in GGBS-MgO and PC
97 binders, the leaching properties of the treated soils exposed to the acid rain would be
98 different. Therefore, it is necessary to comprehensively evaluate the leaching behavior
99 of GGBS-MgO stabilized heavy metal-contaminated soils under different acidic
100 conditions: strongly acidic condition (pH = 2.0), moderate acidic condition (pH = 4.0)
101 and neutral condition (pH = 7.0).

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

110

111 **2 Materials and Testing Methods**

112 **2.1 Materials and sample preparations**

113 Kaolin clay is used as a base soil due to its uniform composition (low organic
114 content, homogeneity and uniform mineralogy) and low cation exchange capacity
115 [1-3, 14]. The basic physiochemical properties of the kaolin clay are summarized in

116 **Table 1.** The pH is measured per [ASTM D4972 \[25\]](#) using a pH meter HORIBA D-54.
117 The specific gravity is measured per [ASTM D5550 \[26\]](#). The Atterberg limits are
118 measured per [ASTM D4318 \[27\]](#). The kaolin clay is classified as lean clay (CL) based
119 on the Unified Soil Classification System [\[28\]](#). The moisture content is measured as
120 per ASTM D2216 [\[29\]](#). The grain size distribution is measured using a laser particle
121 size analyzer Mastersizer 2000.

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

131 Pb is used in this study because it is a very common toxic heavy metal in
132 contaminated soils [\[3, 14, 31\]](#). Lead nitrate ($\text{Pb}(\text{NO}_3)_2$) powder (Chemical Analytical
133 Reagent, Sinopharm Chemical Reagent Co., Ltd.) is dissolved in distilled deionized
134 water (DDW) as stock solutions with predetermined Pb concentrations. The simulated
135 acid rain (SAR), used as the extraction liquid (leachant) in the semi-dynamic leaching
136 test, is prepared by diluting nitric acid (HNO_3) and ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) in
137 the DDW. Prior to adding HNO_3 , ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) solution is added to

138 the DDW until the concentration of the sulfate ion (SO_4^{2-}) reaches 0.001 mol/L [2].
139 The stock solutions of SAR are adjusted to three pH value of 2.0, 4.0, and 7.0
140 respectively. SAR with pH of 2.0 represents a strong acid rain in the field [2].

141 Previous studies show that a binder with 9: 1 ratio of GGBS to MgO (dry weight
142 basis) yields relatively higher strength and lower leachability of stabilized
143 contaminated soils [14]. Therefore, the binder consisting of 90% GGBS and 10%
144 MgO (dry weight basis) is prepared. Three binder contents are set as 12%, 15%, and
145 18% (dry weight soil basis) which are typical contents in engineering projects [2]. The
146 water content and the Pb concentration are set as 45% and 2% (i.e., 20000 mg/kg)
147 (dry weight soil basis) to simulate a heavily contaminated site soil [11, 21],
148 respectively. Six mixtures are investigated in total and denoted as GM_iPb_j , where $i =$
149 content of the GGBS-MgO binder (i.e., 12, 15 or 18), and $j =$ Pb concentration (% , 0
150 or 2).

151 The kaolin clay, GGBS and MgO powders are placed in a plastic bottle and are
152 thoroughly mixed by a bench-top mixer. Then the predetermined volume of $\text{Pb}(\text{NO}_3)_2$
153 stock solution is added to the plastic bottle and further mixed for 30 minutes by the
154 mixer to sufficiently homogenize the mixture. The mixture is filled into a cylindrical
155 PVC mold ($\Phi 50 \times H 100$ mm) in five equal height layers. The mold is vibrated
156 manually after each filling to eliminate air bubbles. After five fillings, the mixture is
157 cured under the standard condition ($20 \pm 2^\circ\text{C}$, relative humidity = 95%) for 28 days.
158 In addition, the GGBS-MgO cement paste (GGBS : MgO = 9:1, water : cement = 0.6)
159 is prepared following the same procedure but without adding kaolin clay and

160 $\text{Pb}(\text{NO}_3)_2$ solution. Totally six identical soil samples are prepared with four samples
161 subjected to the semi-dynamic leaching test, and two samples used for the
162 measurement of specific gravity, water content and density before and after the
163 semi-dynamic leaching test. The crushed and sieved sample was also used for ANC
164 and XRD tests. In addition, one GGBS-MgO paste sample is prepared for XRD test.

165 In the authors' previously studies [2, 14], contaminated soils were prepared by
166 spiking clean soil with heavy metal solution at controlled water content, and cured
167 under controlled condition ($20 \pm 2^\circ\text{C}$, relative humidity = 95%) until chemical
168 equilibrium between soil and heavy metal is achieved. The mixture is then thoroughly
169 mixed with binder with designed dosage, compacted under controlled dry density and
170 water content, and cured before subjected to various tests. The soil sample preparation
171 method presented in this study is more time effective but the chemical reaction
172 between Pb and kaolin may not achieve equilibrium, which may influence the
173 leaching characteristic and microstructural properties of the stabilized soils. Further
174 study is warranted to address this aspect.

175

176 **2.2 Testing Methods**

177 The semi-dynamic leaching test is conducted as per ASTM C1308-08 [32]. Four
178 replicate samples are tested with three different extraction leachants with pH = 2.0,
179 4.0, and 7.0, respectively. The ratio of the liquid volume to the solid superficial area is
180 $9.5 \text{ (mL/cm}^2\text{)}$. The leachant is replenished at 2 h, 7 h, 1 d and then daily until 11 d. It
181 is noted that the semi-dynamic leaching test is not conducted for the untreated soil as

182 a preliminary test shown that the untreated soil specimen disintegrated immediately
183 after soaking in the leachant with pH of 7.0 (i.e., DDW).

184 The pH value of the leachate before each replenishment is measured using a pH
185 meter HORIBA D-54. An aliquot of the leachate is filtered through a 0.45 μm filter
186 and acidified to $\text{pH} < 2.0$ and the concentration of Pb is measured by inductively
187 coupled plasma optical emission spectrometry (ICP-OES, PerkinElmer Optima 8000).
188 Triplicate measurements of pH and Pb concentration are conducted for each sample
189 and the averaged values are reported. The coefficient of variation (COV) values of the
190 pH and Pb concentration for the triplicate measurements are $< 3\%$ indicating the good
191 repeatability of the results. The dry density of each sample is calculated from the
192 measured water content and density of two reduplicate stabilized soils before and after
193 the semi-dynamic leaching test.

194 The cumulative mass of leached Pb is calculated by the following equation:

$$195 \quad A_{i,\text{Pb}} = \sum c_i \times V_i \quad (1)$$

196 where $A_{i, pb}$ = the cumulative mass of leached Pb after i th leaching (mg), c_i = the
197 concentration of Pb after i th leaching (mg/L), and V_i = the volume of the leachate (L),
198 which is 1.66 L in this test. The cumulative fraction of leached mass at time t (CFL) is
199 calculated by:

$$200 \quad CFL = \frac{A_{i,\text{Pb}}}{m} \quad (2)$$

201 where m = the total mass of Pb in the specimen (mg). The observed diffusion
202 coefficient (D^{obs}) is calculated using the following equation:

203
$$D^{\text{obs}} = \frac{\pi}{4} \left(\frac{CFL}{\sqrt{t}} \cdot \frac{V}{S} \right)^2 \quad (3)$$

204 where V = volume of the specimen (cm^3), S = surface area of specimen (cm^2) and t =
 205 leaching time (s). Herein, D^{obs} is a retarded observed diffusion coefficient since it
 206 represents both diffusive and sportive properties of the soils [3, 33].

207 ANC test is performed according to the procedures developed by [Stegemann and](#)
 208 [Côte \[34\]](#). Approximately 10 g soil is sampled from the hand broken sample cured
 209 under the standard condition for 28 days, crushed, sieved ($< 100 \mu\text{m}$), and mixed with
 210 100 mL distilled water. A series of titration tests are conducted on the soil-distilled
 211 water mixture using 0.1 M nitric acid as the extraction liquid. An automatic titrating
 212 device (Auto Titrator ZDJ-4A) is used to fill extraction liquid until the leachate pH of
 213 mixture achieves the target value. Approximately 10 mL leachate after each titration
 214 test is collected and filtered through a $0.45 \mu\text{m}$ filter, and then the concentration of Pb
 215 measured by ICP-OES (PerkinElmer Optima 8000). The COV value of the added acid
 216 volume less than 6%. This test is performed in duplicate and the average results are
 217 reported.

218 The slope of the titration curve (i.e., acid added to the soil versus leachate pH is
 219 expressed as an index of β by [Yong \[35\]](#):

220
$$\beta = - \frac{dC_A}{dpH} \quad (4)$$

221 where dC_A (cmol) = the increment of moles of acid added to the soil.

222 After the semi-dynamic leaching test, one specimen is used for MIP test
 223 conducted as per ASTM D4404 [36]. The MIP test is used to determine the pore size

224 distribution of the soil-binder mixture. Approximate 1 cm³ soil sample is collected
225 from the specimen's surface to the same depth by carefully cutting with a stainless
226 steel knife. Then the collected samples are frozen by the liquid nitrogen (boiling point
227 is -195°C). The frozen samples are dried in a vacuum chamber under -80°C. The MIP
228 tests are performed on dried samples using an Auto Proe IV 9510 mercury intrusion
229 porosimeter. The pore diameter is calculated using the following capillary pressure
230 equation according to ASTM D4404 [36]:

$$231 \quad d = -\frac{4\tau \cos \alpha}{p} \quad (5)$$

232 where d (μm) = pore diameter; τ (N/m) = the surface tension; α (°) = contact angles of
233 mercury with the solid; and p (N/m²) = applied pressure of mercury intrusion. In this
234 study, the contact angle is set as 139° and surface tension value is set as 4.84×10⁻⁴
235 N/mm.

236 The XRD tests are performed on samples of GGBS-MgO paste and 18%
237 GGBS-MgO stabilized kaolin spiked with 2% Pb that are cured under the standard
238 condition for 28 days. Prior to the XRD analysis, 10 g sample is collected, air dried,
239 ground, and sieved (< 0.075 mm), and frozen using liquid nitrogen (-195°C) to be
240 dried by sublimation of the frozen water at -80°C. The XRD test is conducted on
241 gold-coated samples on RigakuD/Max-2500 using a Cu-Kα source with a wavelength
242 of 1.5405 Å. The instrument is operated at 40 kV and 20 mA. A step size of $2\theta = 0.02^\circ$
243 and a scanning speed of 5 s/step are used in the step scan mode. Samples are analyzed
244 over a range of 2θ from 10° to 50°. The binder content, curing time, Pb concentration
245 and number of replicate samples for the various tests are summarized in **Table 4**.

246

247 **3 Results and Analyses**

248 **3.1 Dry Density**

249 **Table 5** shows the properties of the Pb-contaminated kaolin clay treated by different
250 contents of GGBS-MgO before the semi-dynamic leaching test. The water content
251 and porosity slightly decrease with increasing content of GGBS-MgO, whereas dry
252 density values are practically the same regardless of GGBS-MgO content. **Tables 6**
253 **and 7** present the dry density and normalized dry density after leaching under
254 different SAR pH conditions. It is shown that the change of both dry density and
255 normalized dry density are insignificantly with GGBS-MgO content or SAR pH.

256

257 **3.2 Cumulative Fraction Leached and Leachate pH**

258 **Fig. 1** shows the evolution of the cumulative Pb fraction (*CFL*) and leachate pH
259 with time for samples with different GGBS-MgO contents under SAR pH = 2.0, 4.0,
260 and 7.0 . It can be seen that the *CFL* gradually increase throughout the entire leaching
261 time. When GGBS-MgO content increases from 12 to 18%, *CFL* decreases regardless
262 of the SAR pH. The binder content only has a marginal influence on the *CFL* when it
263 exceeds 15%. At the same binder content and time, the increments of *CFL* are much
264 more significant when SAR pH decreases from 4.0 to 2.0 than those when SAR pH
265 decreases from 7.0 to 4.0. The observation is consistent with those reported by [2]
266 where PC is used as the binder for stabilizing Pb-contaminated kaolin soil. When the

267 SAR pH values are 4.0 or 7.0, the leachate pH curves are close to each other and both
268 are approximately 10.5. When the SAR pH decreases from 4.0 to 2.0, a remarkable
269 decrease of the leachate pH to ~2.5 is observed because the amount of the alkaline
270 hydration products formed in the soil matrix is not sufficient to buffer the acid
271 solution.

272

273 **3.3 Observed diffusion coefficient**

274 The cumulative Pb per cross-section area of the soil is plotted against $\log(t)$ in
275 **Fig. 2** for different SAR pH values. The slopes of the regression lines are calculated in
276 **Table 8**. According to USEPA (Method 1315), if the slope is close to 1 (slope > 0.65),
277 surface dissolution will be the dominant leaching mechanism. If the slope is close to
278 0.5 ($0.35 < \text{slope} \leq 0.65$), diffusion is the leaching mechanism. If the slope is lower
279 than 0.35, wash-off occurs (or depletion if it is found in the middle or at the end of the
280 test). This study shows that for SAR pH = 2.0, the leaching mechanism is dissolution
281 while it is diffusion for SAR pH = 4.0 and 7.0. The observed diffusion coefficients
282 (D^{obs}) are in the range of 10^{-18} - 10^{-12} m²/s, which agree with the results from previous
283 studies [37-41]. The comparisons of D^{obs} for Pb in GGBS-MgO, fly ash,
284 quicklime-sulfate and PC stabilized soils [37-42] are shown in **Fig. 3**. Only a few D^{obs}
285 values exist at leachant pH 2.0 due to the strong dissolution effect. When SAR pH is
286 4.0 or 7.0, D^{obs} decreases as the GGBS-MgO content increases from 12% to 18%. The
287 main hydration products in the GGBS-MgO binder are calcium silicate hydrate
288 (C-S-H), hydrotalcite-like phases (Ht) and brucite (Mg(OH)₂) if there is excess MgO

289 [20]. The formation of brucite and Ht causes a large solid volume expansion and fill
290 pores in stabilized soils [20], leading to more compacted soil structure [2] and
291 therefore lower D^{obs} with higher binder dosages. When the SAR pH decreases from
292 7.0 to 4.0, the more aggressive SAR attack on the hydration products leads to their
293 gradual dissolution and consequently higher porosity (see “MIP test” section),
294 resulting in slightly higher D^{obs} .

295

296 3.4 Acid neutralization capacity

297 **Fig. 4(a)** illustrates the titration curves for Pb-contaminated kaolin clay treated
298 by GGBS-MgO. Before adding the acid, the soil with 12% GGBS-MgO binder
299 displays a slightly lower leachate pH than that of the soil with 18% GGBS-MgO
300 binder (i.e., soil pH = 9.9 versus 10.3). Higher GGBS-MgO binder content increases
301 the initial soil pH and thereby could increase the resistance against acid attack. **Fig.**
302 **4(b)** shows the values of β computed by **Eq. (4)**. Values of β gradually decrease with
303 leachate pH dropping from 10.0 to 5.0. The leachates with pH lower than 5.0 are not
304 appropriate for evaluating the buffering capacity of the contaminated kaolin in this
305 study since a certain amount of soil minerals might have dissolved in the ANC test
306 [35]. **Du et al. [14]** stabilized Zn and Pb contaminated soils with a phosphate-based
307 binder and observed the turning point of β occurs at the leachate pH of 5.0. In **Fig. 4(b)**
308 the turning point of β is around 7.0 which is slightly higher than **Du et al. [14]** because
309 the kaolin clay used in this study has less organic component and therefore less acid
310 buffer capacity than natural clay used by **Du et al. [14]**. **Fig. 4(c)** shows the variation

311 of the leached Pb concentration ($\mu\text{g}/\text{kg}$ dry soil) with leachate pH obtained from the
312 ANC test. When the leachate pH is in the range of 2.0 to 4.0, leached Pb
313 concentration decreases noticeably with the increasing GGBS-MgO content or pH. In
314 contrast, leached Pb concentration is lower than $0.015 \mu\text{g}/\text{kg}$ and the values are
315 practically the same when the leachate pH ranges from 5.0 to 10.0 regardless of the
316 GGBS-MgO content or leachate pH.

317

318 **3.5 Pore size distribution**

319 **Fig. 5** presents the cumulative pore volumes for the Pb-contaminated kaolin clay
320 treated by 12% and 18% GGBS-MgO under different pH conditions. Under the same
321 SAR pH condition, the specimens stabilized with 12% GGBS-MgO have notably
322 larger cumulative pore volume than the specimens stabilized with 18% GGBS-MgO.
323 At the same GGBS-MgO content, the cumulative pore volume decreases as the SAR
324 pH increases, which is more noticeable when SAR pH decreases from 4.0 to 2.0.

325 **Fig. 6** shows the pore volumes for pore diameters in different ranges: $< 0.01 \mu\text{m}$
326 (intra-aggregate), 0.01 to $10 \mu\text{m}$ (inter-aggregate) and $> 10 \mu\text{m}$ (air pores) respectively.
327 This classification of pore sizes is suggested by [Horpibulsuk et al. \[43\]](#) for the cement
328 and fly ash-stabilized silty clay. The volume percentages of above classified pores are
329 shown in **Table 9**. Regardless of the GGBS-MgO content, the proportions of air pores
330 and intra-aggregate pores increase (more noticeable for air pores), whereas those of
331 inter-aggregate pores decrease when the SAR pH decreases from 7.0 to 2.0.
332 Increasing GGBS-MgO content is found to decrease the proportions of air pores while

333 increase those of inter- and intra-aggregate pores.

334

335 **3.6 X-ray diffraction analysis**

336 XRD tests are conducted on the GGBS-MgO paste and 18% GGBS-MgO
337 stabilized kaolin spiked with 2% Pb to investigate the emerging reaction products in
338 the GGBS-MgO paste and stabilized Pb-contaminated kaolin. The results are
339 presented in **Fig 7**. For GGBS-MgO paste samples, the characteristic peaks of Ht at
340 $2\theta \approx 11.5^\circ$ and 22.9° agree well with the findings of other researchers [20, 44]. In
341 addition, C-S-H has been detected at $2\theta \approx 29.8^\circ$, 31.6° , 38.0° and 48.7° . The C-S-H has
342 lower ratio of calcium and silicon, and therefore its peak is close to the calcite (CaCO_3)
343 ($2\theta = 29.8^\circ$) as reported by previous studies [20, 44]. MgO is identified suggesting
344 that it has not been fully consumed after 28 days of curing. For Pb-contaminated
345 kaolin treated with 18% GGBS-MgO, the 2θ values of the C-S-H and Ht are found at
346 $\sim 31.7^\circ$ and 11.3° respectively. The characteristic peak of quartz (SiO_2) has been
347 detected at $2\theta \approx 33.6^\circ$. The 2θ of the kaolinite is detected at $2\theta \approx 12.6^\circ$, 19.8° and 34.8° .
348 Additionally, a trace peak of lead carbonated hydroxide hydrate (hydrocerussite,
349 $\text{Pb}_2(\text{CO}_3)_2(\text{OH})_2$) has been detected at $2\theta = 34.2^\circ$, which agrees well with Jin and
350 Al-Tabbaa (2014a) [20] that the main immobilization mechanism for Pb by
351 GGBS-MgO binder is the formation of hydrocerussite.

352

353 **4 Discussion**

354 This study demonstrates that the SAR pH and GGBS-MgO content affect

355 considerably the leachability and D^{obs} of Pb and microstructural properties of the
356 GGBS-MgO stabilized Pb-contaminated kaolin clay. The mechanisms controlling the
357 variation of these features are summarized as follows:

358 (1) As the SAR pH decreases from 7.0 to 2.0, the hydration products (C-S-H and
359 Ht) and kaolin might have been gradually dissolved although it is not explored in this
360 study. The treated Pb-contaminated soils subjected to the acidic conditions (pH 4.0 to
361 2.0), therefore, possess looser structures as compared to those subjected to the neutral
362 condition (pH 7.0) (**Fig. 6**). As the GGBS-MgO content increases from 12% to 18%,
363 higher amounts of hydration products have formed in the soil matrix [15] which in
364 turn results in lower leached Pb concentration observed in the ANC test (**Fig. 4(c)**)
365 and reduces the cumulative volume of pores and the proportions of air pores obtained
366 from the MIP analyses (**Fig. 5**). More compact structure leads to lower D^{obs} and CFL
367 values of Pb in the stabilized soils [2].

368 (2) Regarding the relative variations of the pore volumes in different sizes
369 (**Table 9**), it is proposed that: (a) acid attack results in disintegration of the soil-binder
370 aggregates because kaolin particles and hydration products filling the intra-aggregate
371 pores dissolve, and therefore the intra-aggregate pores volume is elevated; (b) the
372 aggregates themselves, which are formed by kaolin-cement clusters, gradually and
373 partially dissolve due to the acid attack, resulting in the transformation of
374 inter-aggregate pores to air pores. Hence, the inter-aggregate pores volume decreases
375 with decreasing SAR pH, whereas the air pores volume increases with decreasing
376 SAR pH.

377 (3) Different SAR pH conditions and GGBS-MgO contents affect the soil acid
378 buffering capacity (**Fig. 4**). The β values decreases with decreasing pH because the
379 more hydration products are dissolved with more free hydrogen ions (H^+) in the
380 extraction liquid. As the GGBS-MgO content increases from 12% to 18%, a higher
381 concentration of free hydroxyl ions (OH^-) are produced in the soil pores due to cement
382 hydration, resulting in higher β values.

383 (4) Both soil structure and acid buffering capacity affect CFL and D^{obs} of Pb. The
384 above analyses show that soils exhibit loose structures and low β values when SAR
385 pH or GGBS-MgO content reduces, which in turn results in elevated CFL values (**Fig.**
386 **1**). **Eq. (3)** shows that D^{obs} obtained from the semi-dynamic test has a square
387 relationship with CFL . Consequently, D^{obs} values increase with the decreasing SAR
388 pH or GGBS-MgO content (**Table 8**). At a strong acidic condition (pH 2.0), leaching
389 of Pb is controlled by the mineral dissolution process and therefore D^{obs} is not
390 available.

391 It should be noted that the stabilized soils are cured for 28 d in this study, while it
392 is demonstrated that the mechanical properties/microstructure of GGBS-MgO
393 improve significantly in the long term (> 90 days) [15, 45]. Therefore longer curing
394 time is warranted to fairly evaluate the performance of GGBS-MgO stabilized
395 contaminated soils. Moreover, the tested soil samples are prepared under
396 well-controlled laboratory conditions, so it is suggested that field contaminated soils
397 could be adopted in future studies.

398

399 5 Conclusions

400 This study investigates the effect of acid rain with different pH values on the
401 leaching properties of GGBS-MgO stabilized Pb-contaminated kaolin clay via a series
402 of semi-dynamic leaching tests. The effects of the pH of simulated acid rain (SAR)
403 and GGBS-MgO content on the cumulative fraction leached (CFL), observed
404 diffusion coefficient of Pb and pore size distribution profiles of the soils are
405 investigated. Based on the results obtained from this study, the following conclusions
406 can be drawn:

407 (1) The changes of dry density and normalized dry density of Pb-contaminated
408 kaolin clay with GGBS-MgO content or acid rain pH are insignificant within the
409 test conditions in this study.

410 (2) The *CFL* of Pb is notably affected by the simulated acid rain pH and
411 GGBS-MgO content. *CFL* decreases with increasing GGBS-MgO content while
412 increases with decreasing pH, and its increment is more notable at pH 2.0. At pH
413 2.0, mineral dissolution is found to be the dominant process that controls the
414 leaching of Pb.

415 (3) The dominant leaching mechanism of Pb is diffusion at pH 4.0 and 7.0. D^{obs}
416 values of Pb decrease with increasing GGBS-MgO content. The results obtained
417 from this study and those reported in the literature demonstrate that D^{obs} values
418 of Pb increases with decreased pH of leachant tested in the leaching tests.

419 (4) The Pb-contaminated soils with higher GGBS-MgO content display flatter acid
420 neutralization capacity titration curves and higher acid buffer capacity index.

421 When the leachate pH ranges from 2.0 to 4.0, the leached Pb concentration
422 obtained from the ANC test decreases noticeably with increasing GGBS-MgO
423 content or leachate pH, while it changes insignificantly regardless of
424 GGBS-MgO content or leachate pH when pH is in the range of 5.0 to 10.0.

425 (5) MIP results show that the cumulative pore volume decreases as the simulated
426 acid rain pH or GGBS-MgO content increases. The volume percentages of air
427 pores and intra-aggregate pores increase while that of inter-aggregate pores
428 decreases with decreased SAR pH. Increasing GGBS-MgO content decreases the
429 volume percentages of air pores while increases those of both inter- and
430 intra-aggregate pores.

431 (6) Lead is primarily precipitated as hydrocerussite ($\text{Pb}_2(\text{CO}_3)_2(\text{OH})_2$) in the soil
432 matrix after GGBS-MgO treatment. The acid buffer capacity and pore structure
433 obtained from the ANC test and MIP analyses are essential for interpreting
434 effects of leachant pH and GGBS-MgO content on the leachability and diffusive
435 properties of Pb in the soils tested.

436

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438

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447

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593

594

Table Captions

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

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

597 **Table 3.** Chemical compositions of the kaolin soil, GGBS and MgO used in this study

598 measure by XRF

599 **Table 4.** Binder content, curing time, Pb concentration and number of replicated

600 samples for various tests used in this study

601 **Table 5.** Properties of the stabilized soil before the semi-dynamic leaching tests

602 **Table 6.** Dry density of samples calculated from the measured density and water

603 content immediately after the semi-dynamic leaching tests

604 **Table 7.** Normalized dry density of samples calculated from the measured density and

605 water content immediately after the semi-dynamic leaching tests

606 **Table 8.** Calculated D^{obs} values and leaching mechanisms of Pb for the soils tested

607 between 2 h and 11 d

608 **Table 9.** Distribution of the pore volume percentage of the Pb contaminated kaolin

609 treated by GGBS-MgO after semi-dynamic tests

610

611

612

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 to 0.075 mm)	58
Sand (0.075 to 2 mm)	20.5

613

614

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

615 ^aThe alkalinity of the GGBS is defined as the ratio of contents of CaO, MgO, and
616 Al₂O₃ to that of SiO₂

617

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

Chemical composition (<i>wt%</i>)	Kaolin	GGBS	MgO
CaO	0.36	33.08	0.84
Al ₂ O ₃	39.3	17.9	0.38
MgO	0.06	6.02	96.5
K ₂ O	0.21	0.64	0.01
SiO ₂	52.1	34.3	1.09
Fe ₂ O 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

620

621 **Table 4. Binder content, curing times, Pb concentration and number of**
 622 **replicated samples for various tests used in this study**

Test/ Analysis	Binder content, %	Curing time, days	Pb concentration, %	Number of replicate samples
Dry density	12, 15, 18	28	2	2
Semi-dynamic test	12, 15, 18	28	2	4
ANC ^a	12, 18	28	2	1
MIP ^b	12, 18	28	2	1
XRD ^c	18	28	0, 2	1

623 ^aANC= Acid neutralization capacity;
 624 ^bMIP = Mercury intrusion porosimetry;
 625 ^cXRD = X-ray diffraction

626

627

Table 5. Properties of the stabilized soils before the semi-dynamic leaching tests

	^a Specific gravity, G_s	^b Water content, %	Dry density, ρ_d (g/cm ³)	Porosity, n	Saturation degree, S
GM12Pb2	2.61	41.0	1.26	0.519	0.992
GM15Pb2	2.59	39.7	1.26	0.514	0.973
GM18Pb2	2.55	38.2	1.28	0.498	0.982

628

^a ASTM D5550(ASTM 2014)

629

^b ASTM D2216(ASTM 2010)

630

631 **Table 6. Dry density (g/cm^3) of samples calculated from the measured density**
632 **and water content immediately after the semi-dynamic leaching tests**

Sample / pH	pH = 2.0	pH = 4.0	pH = 7.0
GM12Pb2	1.20	1.22	1.23
GM15Pb2	1.25	1.25	1.26
GM18Pb2	1.25	1.25	1.26

633
634

635 **Table 7. Normalized dry density (g/cm^3) of samples calculated from the measured**
636 **density and water content immediately after the semi-dynamic leaching tests**

Sample / pH	pH = 2.0	pH = 4.0	pH = 7.0
GM12Pb2	0.96	0.97	0.98
GM15Pb2	0.99	0.99	1.00
GM18Pb2	0.98	0.98	0.99

637
638

639 **Table 8. Calculated D^{obs} values and leaching mechanisms of Pb for the soils tested**
 640 **between 2 h and 11 d**

	pH	Slope	R^2	Mechanism	D^{obs} (m ² /s)
GM12Pb2	2.0	0.71	0.992	dissolution	-
	4.0	0.51	0.96	diffusion	1.69×10^{-16}
	7.0	0.44	0.997	diffusion	7.67×10^{-18}
GM15Pb2	2.0	0.8	0.989	dissolution	-
	4.0	0.52	0.972	diffusion	4.90×10^{-17}
	7.0	0.58	0.962	diffusion	5.00×10^{-18}
GM18Pb2	2.0	0.98	0.982	dissolution	-
	4.0	0.6	0.98	diffusion	5.07×10^{-17}
	7.0	0.6	0.948	diffusion	3.77×10^{-18}

641

642

643 **Table 9. Distribution of the pore volume percentage of the Pb contaminated**

644 **kaolin treated by GGBS-MgO after semi-dynamic test**

SAR pH	GM12Pb2			GM18Pb2		
	pH = 2.0	pH = 4.0	pH = 7.0	pH = 2.0	pH = 4.0	pH = 7.0
intra-aggregate pores (%)	1.86	1.80	1.77	2.74	2.00	1.76
inter-aggregate pores (%)	90.76	91.18	91.70	91.00	92.15	93.86
air pores (%)	7.38	7.00	6.53	6.26	5.85	4.38

645

Figure Captions

646

647 **Figure. 1.** CFL for Pb and leachate pH for soils with GGBS-MgO contents of (a) 12%;
648 (b) 15%; and (c) 18%

649 **Figure. 2.** The plots of cumulative Pb against $\log(t)$ from the semi-dynamic tests for
650 specimens with GGBS-MgO content of (a) 12%; (b) 15%; and (c) 18%

651 **Figure. 3.** Variation of D^{obs} for lead with leachant pH obtained from this study and
652 previously published studies

653 **Figure. 4.** (a) Acid titration curves; (b) buffer capacity; and (c) leached Pb
654 concentration of the lead contaminated kaolin clay treated by GGBS-MgO

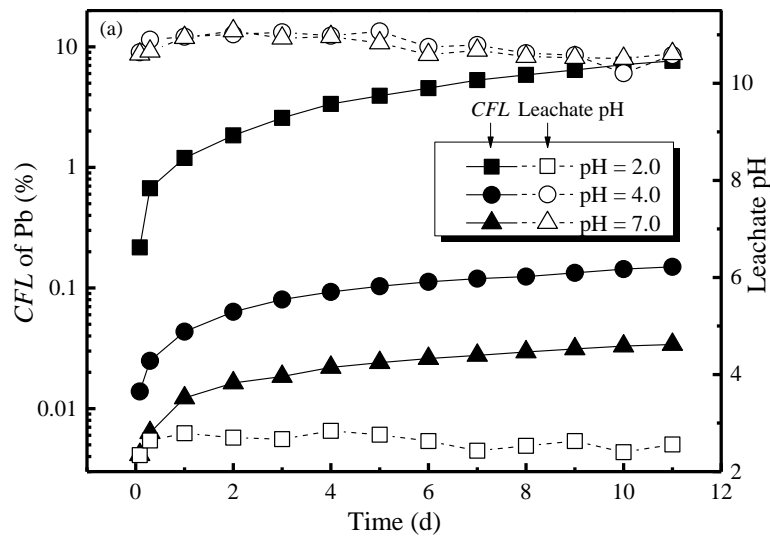
655 **Figure. 5.** Cumulative pore volume of the Pb contaminated kaolin treated by
656 GGBS-MgO after semi-dynamic leaching test

657 **Figure. 6.** Pore volume percentage of the Pb contaminated kaolin treated by
658 GGBS-MgO after semi-dynamic test

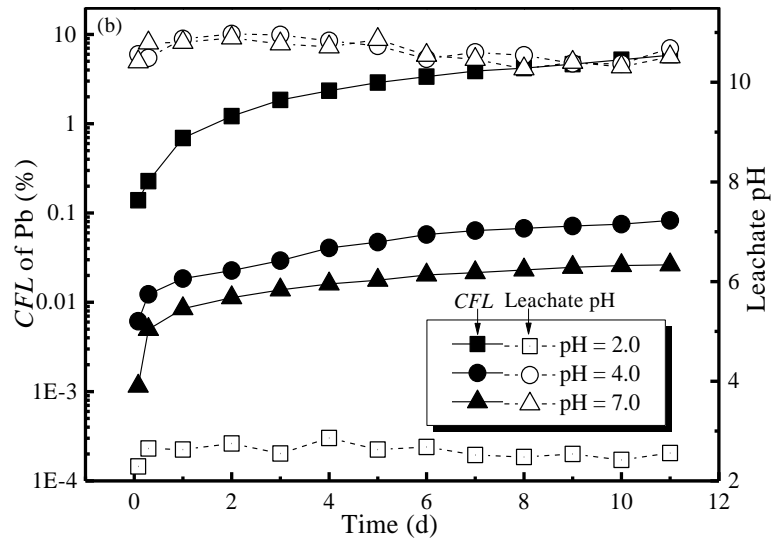
659 **Figure. 7.** X-ray diffractograms of the GGBS-MgO paste and GGBS-MgO treated
660 Pb-contaminated kaolin cured at the normal condition for 28 days

661

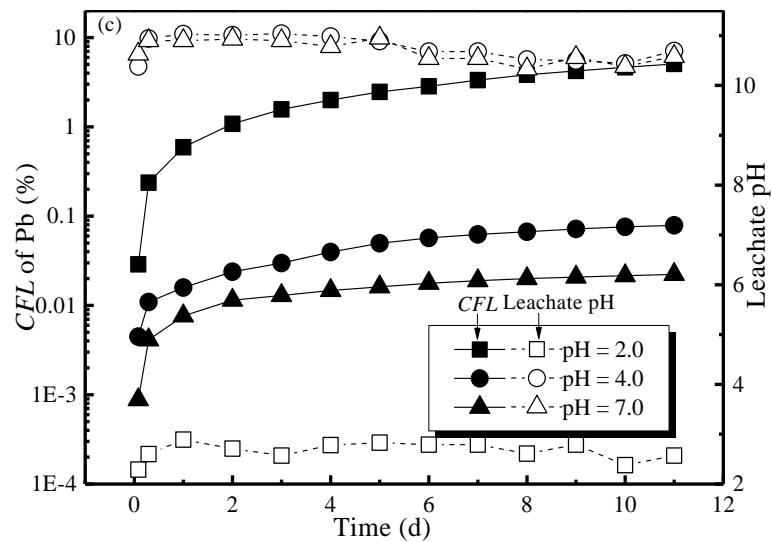
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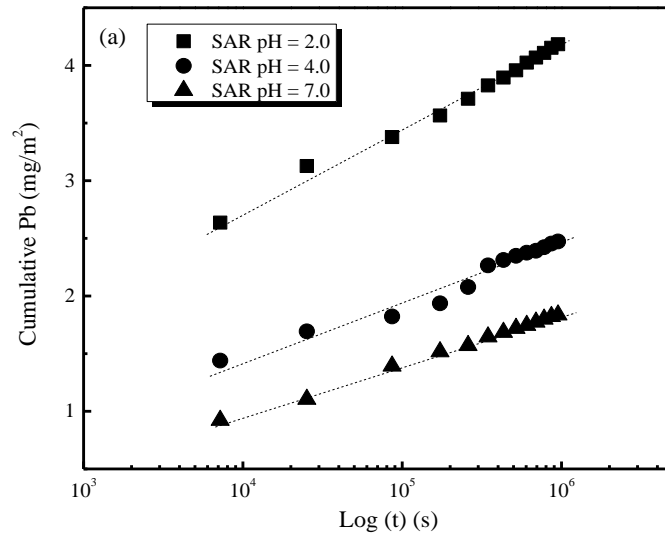
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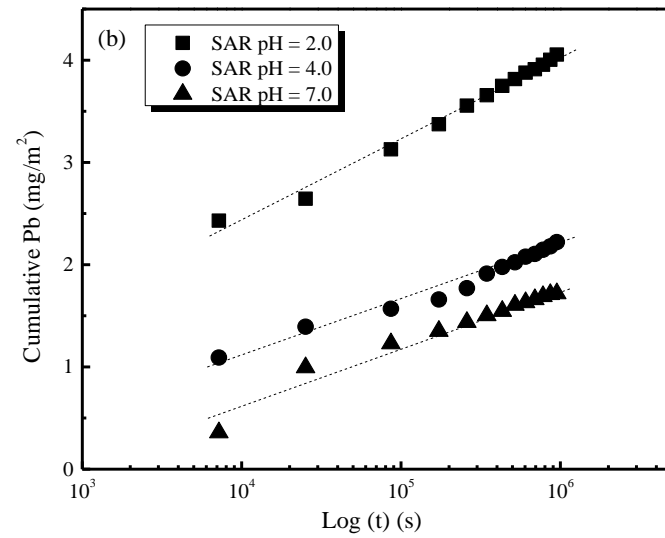
Fig. 1 CFL for Pb and leachate pH for soils with GGBS-MgO contents of (a)

667

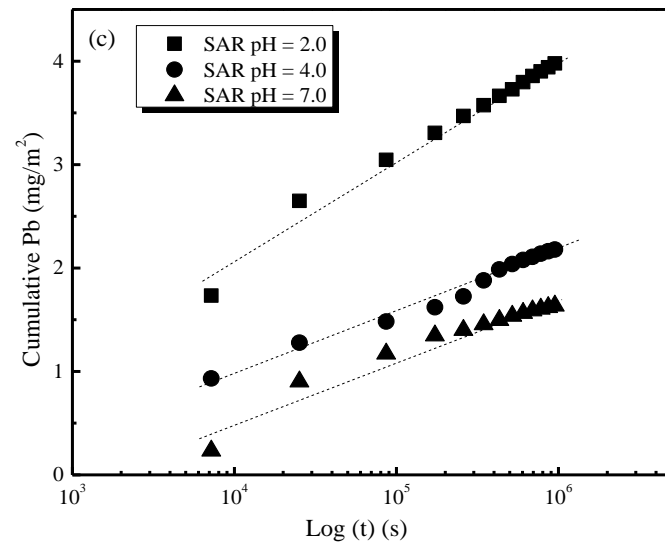
12%; (b) 15%; and (c) 18%.



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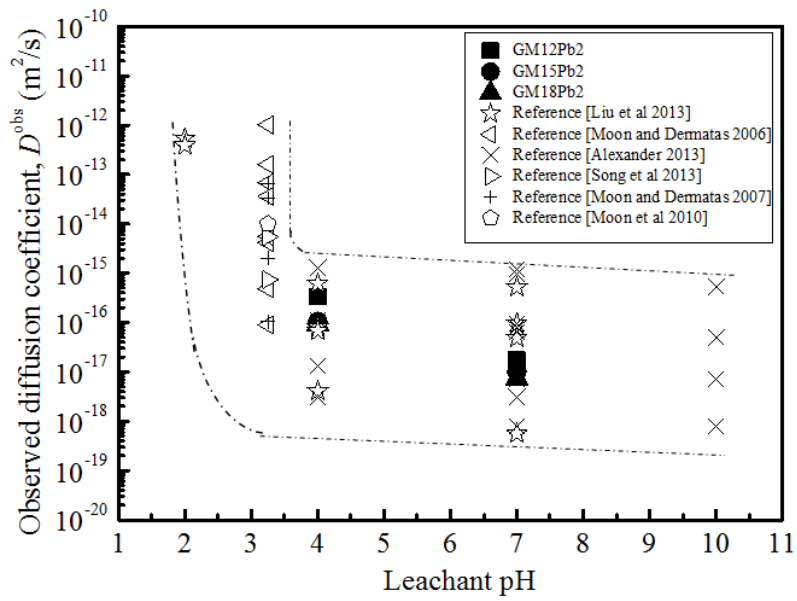
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671 **Fig. 2** The plots of cumulative Pb against log (t) from the semi-dynamic tests for

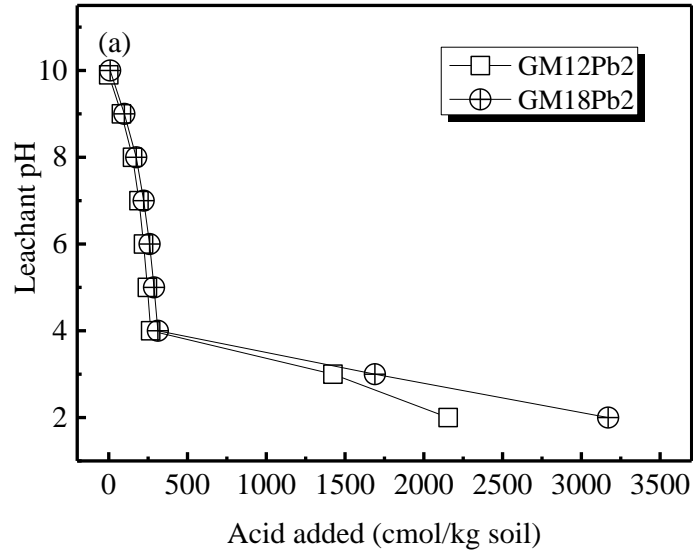
672 specimens with GGBS-MgO content of (a) 12%; (b) 15%; and (c) 18%.



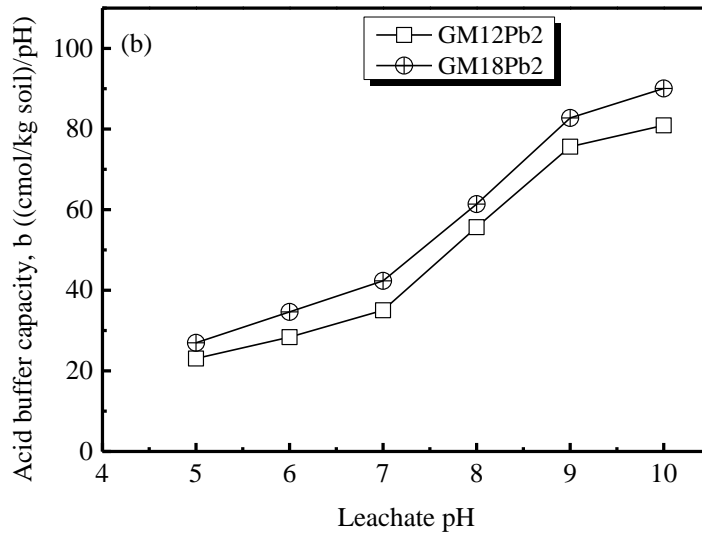
673

674 **Fig. 3 Variation of D^{obs} for lead with leachant pH obtained from this study and**
 675 **previously published studies**

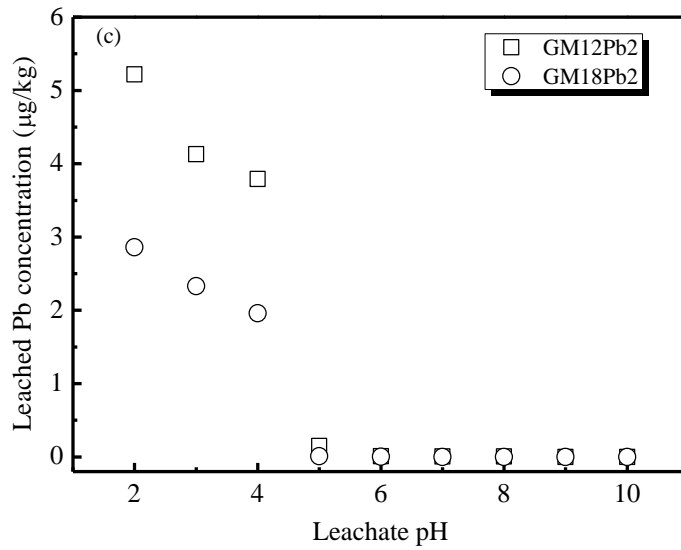
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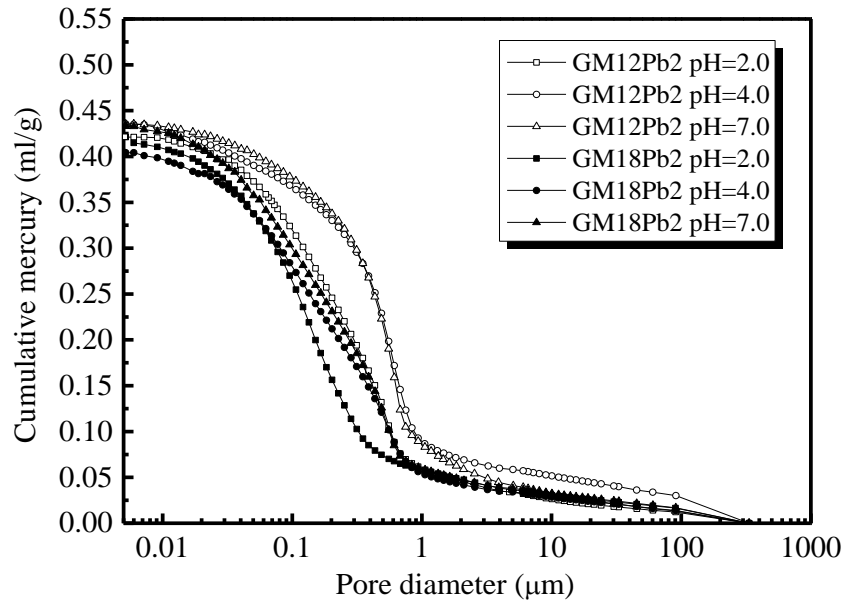
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Fig. 4 (a) Acid titration curves; (b) buffer capacity; and (c) leached Pb

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concentration of the lead contaminated kaolin clay treated by GGBS-MgO



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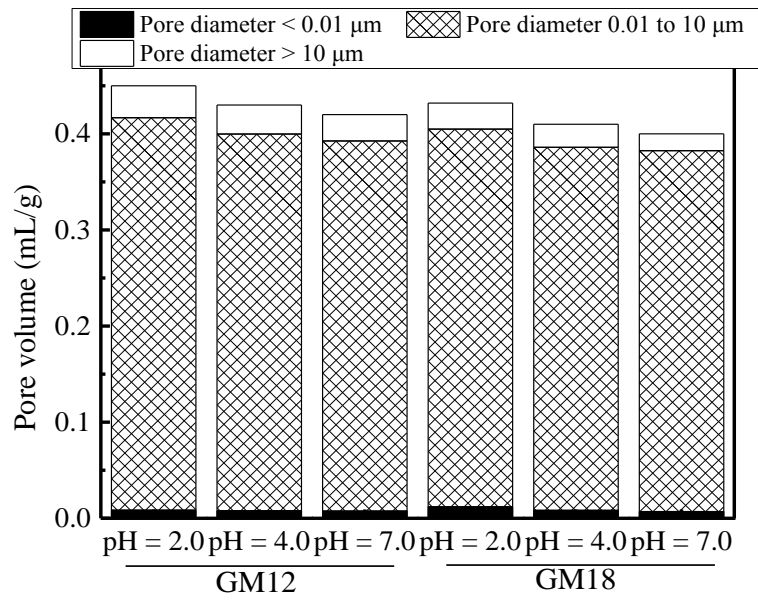
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Fig. 5 Cumulative pore volume of the Pb contaminated kaolin treated by

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GGBS-MgO after semi-dynamic leaching test

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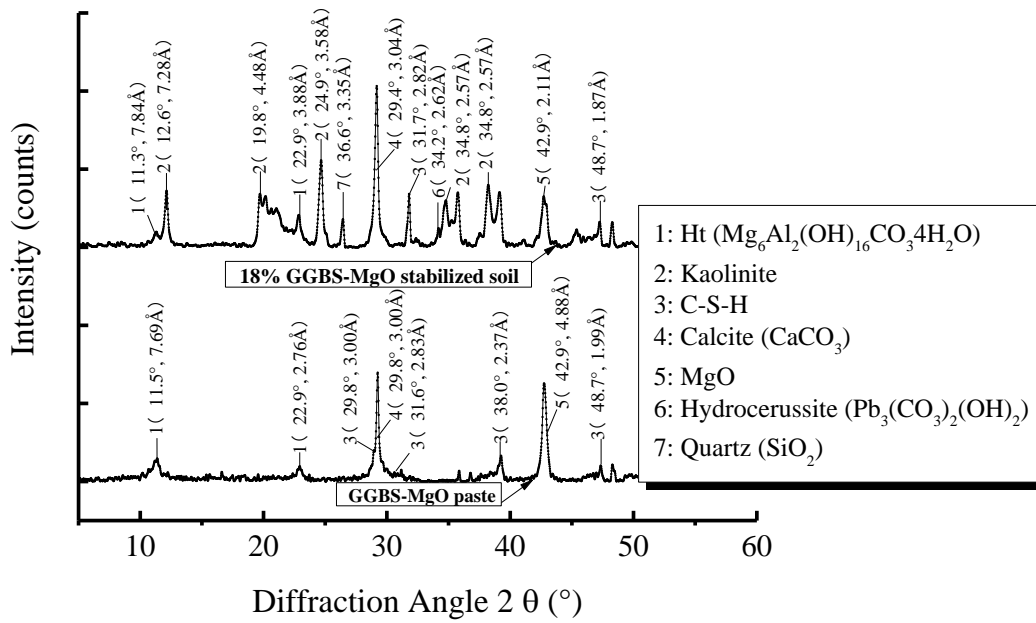
686

687 **Fig. 6 Pore volume percentage of the Pb contaminated kaolin treated by**

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GGBS-MgO after semi-dynamic test

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691 **Fig. 7 X-ray diffractograms of the GGBS-MgO paste and GGBS-MgO treated**

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Pb-contaminated kaolin cured under standard condition for 28 days

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