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1 **Influence of wet-dry cycles on vertical cutoff walls made of reactive**
2 **magnesia-slag-bentonite-soil mixtures**

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Abstract

The strength and hydraulic conductivity of vertical cutoff walls consisting of reactive magnesia-activated ground granulated blast furnace slag (GGBS), bentonite and soil (MSB) have been investigated in previous studies. However, there has been little comprehensive study of the influence of wet-dry cycles on the mechanical and microstructural properties of MSB backfills. In this paper, the durability of MSB backfills when exposed to wet-dry cycles is investigated. The variations in mass change, dry density, pH value, pore size distribution and mineralogy are discussed. The results show that the mass change of Ordinary Portland Cement (OPC)-based and MSB backfills increases with respect to wet-dry cycles. The MSB backfills exhibit up to 8.2% higher mass change than OPC-based ones after ten wet-dry cycles. The dry density, pH value and unconfined compressive strength of MSB backfill decrease with the increasing number of wet-dry cycles. Increasing the GGBS-MgO content from 5% to 10% in MSB backfills results in 2.1 - 2.3 times higher strength, corresponding to a reduction of 2% - 12% in cumulative pore volume; while increasing the bentonite content slightly reduces the strength of MSB mixtures, corresponding to an increase of cumulative pore volume by 4.6% - 7.9%. The hydrotalcite-like phases and C-S-H are the primary hydration products in MSB backfills. Moreover, the continuous wet-dry cycles result in the precipitation of calcite (CaCO_3) and nesquehonite ($\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$).

Keywords: Cutoff wall; reactive MgO-activated GGBS; durability; wet-dry cycles; carbonation

45 1. Introduction

46 The soil-cement-bentonite (SCB) cutoff wall has been widely used worldwide in
47 remediation projects for contaminated sites (Ryan and Day 2002; Opdyke and Evans
48 2005; Ruffing and Evans 2014). Such walls have been mainly used to interrupt the
49 pollution pathway and to isolate the contaminant source from a vulnerable receptor
50 (Joshi et al. 2008; Soga and Joshi 2015; Du et al. 2015; Yang et al. 2018 and 2019). As
51 compared to soil-bentonite (SB) and cement-bentonite (CB) cutoff walls, SCB walls
52 possess adequate strength to carry foundation loads. They have the additional economic
53 merit of the reuse of site-excavated soils (Ryan and Day 2002; Opdyke and Evans 2005).
54 In CB and SCB walls, Ordinary Portland cement (OPC) is the primary cementitious
55 material used, and is associated with intensive CO₂ emissions (0.95 t/t OPC) and
56 consumption of raw materials (Benhela et al. 2013). In recent years, many industrial
57 by-products such as ground granulated blast furnace slag (GGBS) and fly ash have
58 become popular as partial substitutions for OPC in geotechnical applications (Jefferis
59 2012; Lam and Jefferis, 2017; Arulrajah et al. 2017a; Arulrajah et al. 2017b; Wu et al.
60 2019). For example, replacing OPC with GGBS in the cutoff wall backfill has been
61 applied extensively in the UK (Jefferis 2012). It has significant environmental and
62 economic benefits, marginally affects the q_u (sometimes with enhancement), slightly
63 decreases pore water pH and notably decreases hydraulic conductivity in the long term
64 (Wu et al. 2019).

65 Compared to OPC, using reactive MgO as the activator for GGBS has many
66 potential technical benefits, in particular for soil remediation applications. The main

67 hydration products in reactive MgO-activated GGBS are calcium silicate hydrate (C-S-
68 H), hydrotalcite and brucite ($\text{Mg}(\text{OH})_2$, if MgO is excessive) (Du et al. 2016; Jin et al.
69 2015; Jin and Al-Tabbaa 2014a; Wang et al. 2016; Yi et al. 2013). Due to the absence
70 of highly soluble portlandite with a high equilibrium pH value (~ 12.5) and the
71 formation of the above-mentioned products with excellent adsorptive capacities, MgO-
72 GGBS blends have shown superiority over OPC for contaminant immobilization in a
73 number of laboratory studies and field trials (Du et al. 2016; Jin et al. 2015; Jin et al.
74 2016; Jin and Al-Tabbaa, 2014a and b; Wang et al. 2016).

75 Recently, this novel cement has been applied to form an innovative cutoff wall
76 backfill material, together with bentonite and site sandy soil (MSB) (Wu et al. 2019).
77 The unconfined compressive strength (q_u) and hydraulic conductivity (k_w) of the
78 proposed backfill permeated with tap water are in the range of 230 - 520 kPa and
79 1.1×10^{-10} - 6.3×10^{-10} m/s at 90-day-curing, respectively, complying well with the
80 commonly adopted design limits ($q_u \geq 100$ kPa and $k_w \leq 1.0 \times 10^{-8}$ m/s) (ICE 1999; Ryan
81 and Day 2002;). More importantly, this backfill showed excellent durability under
82 exposure to sodium sulfate (Na_2SO_4) or lead-zinc (Pb-Zn) solutions (Wu et al. 2019).
83 Environmental and cost assessments demonstrated that this innovative MSB backfill
84 material reduced CO_2 emissions by 85% with $\sim 15.3\%$ - 16.9% lower cost when
85 compared with OPC-based SCB backfills.

86 In a scenario of cutoff wall installations, the groundwater levels at each side of the
87 barrier may fluctuate over time (Evans 1991; Ross and Beljin 1998). The potential
88 changes in the q_u and k_w of the cutoff wall exposed to cyclic wet-dry actions have been

89 recognized as a medium- to long-term performance concern (Malusis et al., 2011;
90 National Research Council, 2007; Soga and Joshi, 2015). For CB and SCB walls,
91 noticeable cracks were observed due to shrinkage in the drying phase (Soga and Joshi
92 2015), which would lead to the increase of k_w by 2 - 3 orders (Joshi et al., 2009).
93 Previous studies also illustrated that the q_u of MgO-GGBS stabilized kaolin clay
94 reduced dramatically with increasing wet-dry cycles (Du et al., 2016). Thus, it is
95 necessary to evaluate the durability of the innovative MSB backfill under wet-dry
96 cycles.

97 This paper aims to investigate the effect of wet-dry cycles on the durability of
98 innovative MSB backfill. A series of laboratory tests, including mass change, dry
99 density, pH value, unconfined compression tests, Mercury Intrusion Porosimetry (MIP)
100 and X-ray diffraction (XRD) are performed in order to reveal the effects of wet-dry
101 cycles on the properties of MSB backfills.

102 **2. Materials and specimen preparation**

103 2.1 Solid materials

104 The backfill materials were prepared using Nanjing local soil, powdered sodium
105 activated calcium-bentonite, MgO and GGBS. The Nanjing local soil is classified as a
106 clayey sand based on the Unified Soil Classification System (ASTM 2017). The
107 commercial powdered sodium activated calcium-bentonite was provided by the
108 MuFeng mineral processing plant in Zhenjiang, China. GGBS and MgO used in this
109 study were obtained from Nanjing and Jinan, China, respectively. The physicochemical
110 properties for the site soil and bentonite are shown in **Table 1**. The moisture and specific

111 gravity were measured according to ASTM D2216 (ASTM, 2010a) and ASTM D4318
 112 (ASTM, 2010b), respectively. The pH was measured by ASTM D4972 (ASTM, 2018a)
 113 using a pH meter HORIBAD-54. The cation exchange capacity was measured as in
 114 ASTM D7503 (ASTM, 2010c). The specific surface area was measured by nitrogen
 115 adsorption using a Physisorption Analyzer ASAP2020 according to (Cerato and
 116 Lutenegger, 2002). The medium reactivity MgO (reactivity is 102 s, determined by the
 117 acetic acid test according to Shand (2006)) was selected due to its appropriate reactivity
 118 and cost (Wu et al., 2018a and b; Jin and Al-Tababa, 2014c). The chemical
 119 compositions of the soil, GGBS and MgO used for this study are shown in **Table 2**, as
 120 measured by X-ray fluorescence (XRF) (ARL™ SMS-2000).

121 **Table 1. Physicochemical properties of the local sandy-clay and bentonite**

| Index | Values | | | |
|--|-------------|-----------|-------|-------|
| | clayey sand | bentonite | GGBS | MgO |
| Moisture, (%) | 4.81 | 11.2 | - | - |
| pH (liquid to solid ratio at 1) | 7.32 | 8.6 | 10.96 | 10.53 |
| Specific gravity, G_s | 2.62 | 2.66 | - | - |
| Plastic limit, w_p (%) | - | 55 | - | - |
| Liquid limit, w_L (%) | - | 103 | - | - |
| Grain size distribution (%) | | | | |
| Clay (<0.002 mm) ^a | 5.62 | 99 | - | - |
| Silt (0.002-0.075 mm) ^a | 14.18 | 1 | - | - |
| Sand (0.075-2 mm) ^b | 80.20 | - | - | - |
| Specific surface area, SSA (m ² /g) | - | 378.5 | 0.29 | 28.02 |
| Exchangeable cation (cmol/kg) | | | | |
| Ca ²⁺ | | 22.74 | | |
| Mg ²⁺ | | 1.41 | | |
| Na ⁺ | | 53.39 | | |
| K ⁺ | | 0.53 | | |
| Sum | | 78.07 | | |

122 ^a Measured using a laser particle analyzer Mastersizer 2000 (Malvern Instruments Ltd., UK)

123 ^b Measured with standard #10 - #200 sieves

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 125
 126

127 **Table 2. Chemical compositions of the clayed sand, OPC, GGBS and MgO (by**
 128 **wt%) used in this study determined by XRF**

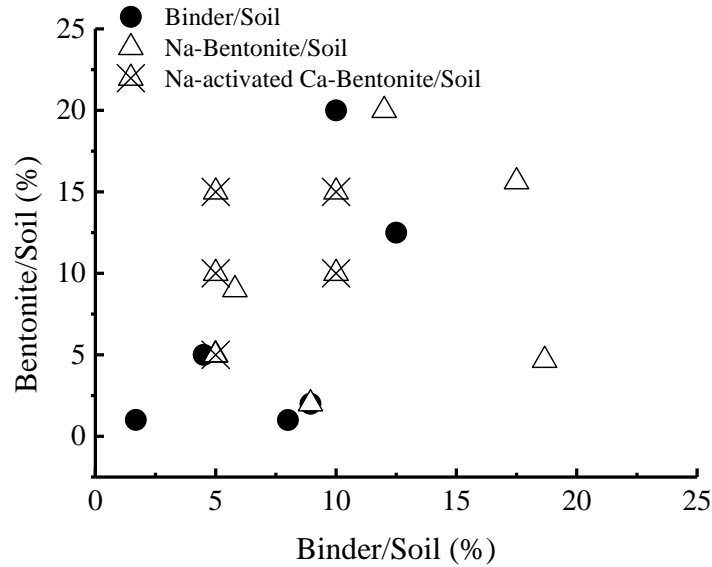
| Oxides | Clayey sand | OPC | GGBS | MgO |
|--------------------------------|-------------|-------|-------|-------|
| CaO | 0.41 | 49.75 | 34.00 | 0.23 |
| Al ₂ O ₃ | 35.76 | 10.87 | 17.90 | 0.28 |
| MgO | 0.06 | 2.26 | 6.02 | 92.95 |
| K ₂ O | 0.15 | 0.75 | 0.64 | 0.01 |
| SiO ₂ | 48.73 | 22.6 | 34.3 | 0.28 |
| Fe ₂ O ₃ | 6.13 | 3.50 | 1.02 | - |
| SO ₃ | 0.07 | 3.84 | 1.64 | 0.45 |
| MnO | 0.11 | 0.24 | 0.28 | 0.01 |
| Loss of ignition (%) | 8.58 | 6.19 | 4.20 | 5.79 |

129

130 2.2 Backfill mix design and specimen preparation

131 **Table 3** shows the mix proportions of the studied backfills. The MgO to GGBS
 132 ratio was 1:9 to achieve good strength and the lowest hydraulic conductivity based on
 133 preliminary studies (Jin et al., 2015; Wu et al., 2018a; Wu et al., 2019). The binder
 134 (OPC or GGBS-MgO) dosages (by weight of dry soil) ranged from 2.5% to 12.5%
 135 based on a review of previous studies (**Fig. 1**) as reported in Wu et al. (2019), which
 136 included both field and lab test results of SCB cutoff walls. Furthermore, since natural
 137 high-quality Na-bentonite in China is scarce, in practice engineers usually prefer to
 138 choose a higher dosage ($\geq 5\%$) of sodium activated calcium-bentonite. The raw solid
 139 materials, including the clayey sand, powdered sodium activated calcium-bentonite,
 140 OPC or MgO and GGBS were weighed and mixed in a 2-L Hobart stainless steel mixer
 141 at 30 rpm for 5 min. It should be noted that the backfill specimens were prepared by
 142 mixing the non-prehydrated bentonite with sandy soil-OPC or sandy soil-GGBS-MgO
 143 mixture. Then the predetermined amount of tap water (pH = 6.8; $EC = 3.3 \mu\text{S}/\text{cm}$) was
 144 added and mixed at 60 rpm for 10 min to achieve homogeneity with the same slump
 145 value ($150 \pm 5 \text{ mm}$) (Wu et al., 2019). In the OPC-based backfill, the content of the

146 GGBS used in this study was 80% replacement of OPC, which was reported to exhibit
 147 the lowest k_w and highest q_u (Opdyke and Evans, 2005). The mixtures were poured into
 148 cylindrical moulds with sizes of $\Phi 50 \times H100$ mm and cured for 90 days under
 149 standard curing conditions ($T = 20 \pm 2$ °C, RH = 95%).



150
 151 **Fig. 1 Summary of bentonite/soil and binder/soil ratios in SCB cutoff wall backfill mix**
 152 **designs reported in previous studies (adapted from Wu et al., 2019)**

153
 154 **Table 3. Mix design for the backfills used in this study (wt%)**

| Category ID | Code | Clayey sand | Bentonite | OPC | GGBS | MgO |
|-------------|---------|-------------|-----------|-----|------|-----|
| C | C5 | 100 | - | 5 | - | - |
| CB | C5B5 | 100 | 5 | 5 | - | - |
| | C10B10 | 100 | 10 | 10 | - | - |
| CSB | CS5B5 | 100 | 5 | 1 | 4 | - |
| | CS10B10 | 100 | 10 | 2 | 8 | - |
| | MS5B5 | 100 | 5 | - | 4.5 | 0.5 |
| | MS5B10 | 100 | 10 | - | 4.5 | 0.5 |
| MSB | MS5B15 | 100 | 15 | - | 4.5 | 0.5 |
| | MS10B10 | 100 | 10 | - | 9 | 1 |
| | MS10B15 | 100 | 15 | - | 9 | 1 |

155

156 3. Laboratory testing program

157 The 90-day cured specimens were subjected to wet-dry cycles according to Kamon
 158 et al. (1993). For each drying cycle, the specimens were stored in an oven (30 °C) for

159 48 h, followed by soaking in the distilled water with a temperature of 20 °C for 24 h.
160 The drying temperature was set as 30 °C to represent the typical average temperature
161 of hot seasons in the southeast regions of China and to minimize the effect of
162 accelerated slag hydration due to temperature rise (Du et al., 2016). The duration of
163 such a wet-dry cycle was three days and it was repeated for ten cycles.

164 After the i^{th} wet or dry cycle, the mass change (MC) of the specimen is calculated
165 by Equation (1):

$$166 \quad MC = (m_0 - m_i) / m_0 \quad (1)$$

167 where m_0 = initial mass of the specimen measured prior to the wet-dry cycles, and
168 m_i = mass of the sample measured immediately after the i^{th} drying cycle.

169 The densities of the specimens were measured in triplicate as in ASTM D7263
170 (2018b), and average values were reported. The specimens were subjected to UCT as
171 in ASTM D4219 (ASTM, 2008) with a strain rate of 1%/min. Fragments from the core
172 of the broken UCT specimens were air-dried, crushed and passed through a 2-mm sieve.
173 Ten grams of the sieved soil and 10 ml of distilled water (water to solid ratio = 1: 1)
174 were poured into a glass container to determine the pore water pH. The pH values of
175 the supernatant were measured by a HORIBA D-54 pH meter and the average value
176 was reported.

177 After the 10th wet-dry cycle, dry specimens were prepared to conduct MIP and
178 XRD tests. Briefly, ~1 cm³ sample was collected from the specimen's core by a stainless
179 steel knife and frozen in liquid nitrogen. The frozen sample was dried in a vacuum
180 chamber at -80°C. The MIP tests were conducted according to ASTM D4404 (2010d)

181 using an Auto Pore IV 9510 mercury intrusion porosimeter to determine the pore size
182 distribution. Prior to XRD analysis, the dry specimens were ground and sieved to <
183 0.075 mm. The XRD spectra were obtained using a RigakuD/Max-2500 spectrometer
184 using a Cu-K α source with a wavelength of 1.5405 Å. The instrument was operated at
185 40 kV and 20 mA. A step size of $2\theta = 0.02^\circ$ and a scanning speed of 5 s/step were used
186 over a range of 2θ from 10° to 50° in the step scan mode.

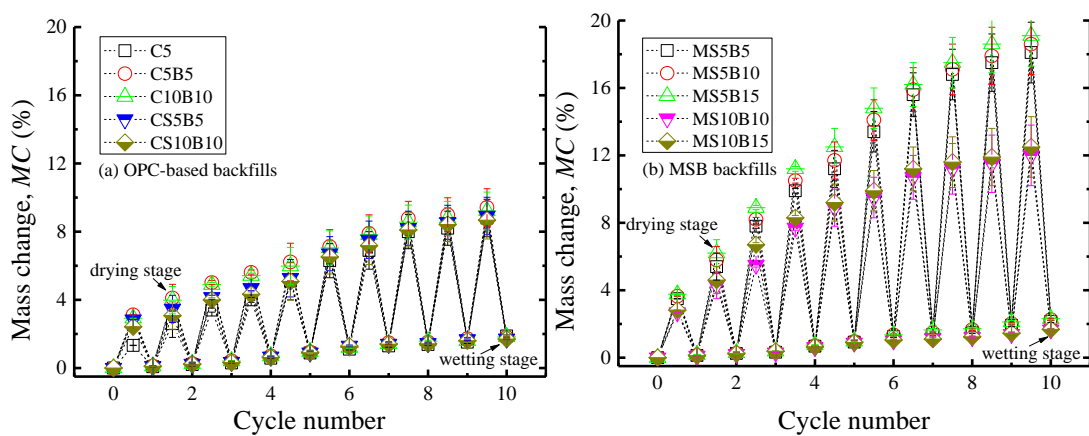
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188 **4. Results and Discussion**

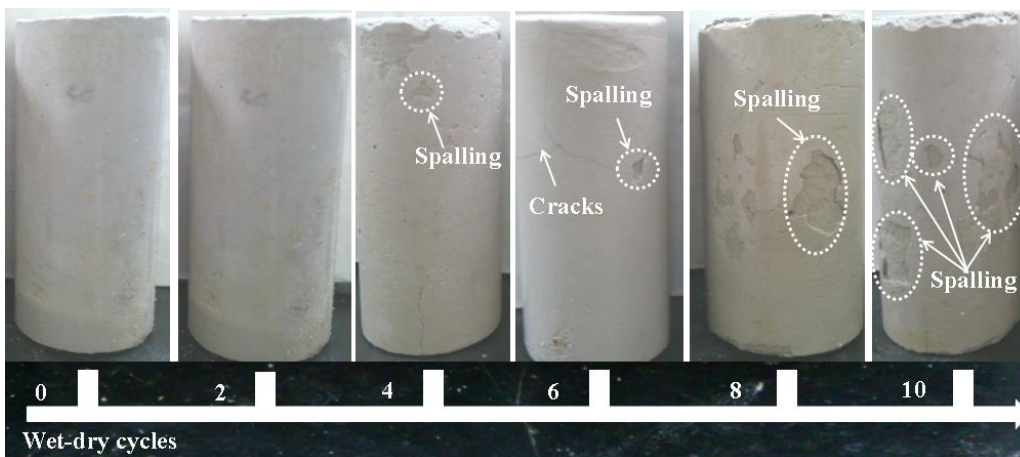
189 4.1 Mass change and visual inspection of physical integrity

190 **Fig. 2 (a)** and **(b)** present the variations in mass change with the wet-dry cycles
191 for the OPC-based and MSB backfills. It can be seen that the mass changes of the two
192 types of backfill increase with respect to the wet-dry cycles. Increasing the binder
193 content (GGBS-MgO) in the MSB backfills reduces mass change (by ~ 9.0 - 11%) (i.e.,
194 comparing MS10B10 and MS10B15 with MS5B10 and MS5B15, respectively). **Fig. 2**
195 **(b)** also indicates that increasing the bentonite content from 5% to 15% leads to a higher
196 mass change for the MSB backfills. This is attributed to the fact that higher bentonite
197 content (i.e., MS5B15) is more sensitive to the loss of water as compared to MS5B5
198 and MS5B10, which would develop matrix suction and increase the shrinkage in the
199 backfills in the drying stages (Nahlawi and Kodikara, 2006; Rowe et al., 2011; Tang et
200 al., 2011). However, the binder and bentonite contents show no significant impact on
201 the OPC-based backfills as shown **Fig. 2 (a)**. After the last wet-dry cycle, the mass
202 changes in MSB backfills reach approximately 12 - 19%, and are slightly higher than

203 those for the OPC-based backfills. Meanwhile, the mass change is much higher in MSB
 204 than OPC at the wetting stage; the difference is much smaller at the drying stage. This
 205 may imply that the water absorption potential of the bentonite in MSB mixtures is better
 206 preserved than that in OPC-based mixtures. It was also found that spalling and macro-
 207 cracks started to develop at the 4th - 6th cycles and were exacerbated with each wet-dry
 208 cycle, as shown in Fig. 3.



209
 210 **Fig. 2 Variations in mass change with wet-dry cycles for (a) OPC-based and (b) MSB**
 211 **backfills**



213
 214 **Fig. 3 Photos showing the physical integrity of MSB backfills during wet-dry cycling**

215 4.2 Dry density

216 **Fig. 4** shows the variations of average values and standard deviations (STD) of
 217 dry densities with wet-dry cycles for OPC-based and MSB backfills. The dry densities

218 of OPC-based and MSB backfills are 1.34 - 1.39 g/cm³ and 1.31 - 1.35 g/cm³ after the
 219 10th wet-dry cycles, respectively. For all backfills, the dry density decreases slightly
 220 with the wet-dry cycles, which is consistent with the mass change evolution as shown
 221 in **Fig. 2**. For the identical binder and bentonite content, it is found that the dry density
 222 of MSB backfills (i.e., M5B5) is higher than that of OPC-based backfills (i.e., C5B5
 223 and CS5B5) after being exposed to ten wet-dry cycles. This may be attributed to the
 224 fact that the C-S-H gels formed in GGBS-MgO have a lower Ca/Si ratio compared to
 225 those formed in OPC, leading to higher shrinkage during the drying process (Du et al.,
 226 2016; Jin et al., 2014b). At the same bentonite content in MSB backfills, the increase
 227 of GGBS-MgO content has only a slight impact on the dry density (i.e., comparing
 228 MS10B10 and MS10B15 with MS5B10 and MS5B15, respectively) whereas the dry
 229 density significantly decreases as the bentonite content increases from 5% to 15% at
 230 the identical GGBS-MgO content (i.e., comparing MS5B5 with MS5B10 and MS5B15)
 231 after ten wet-dry cycles.

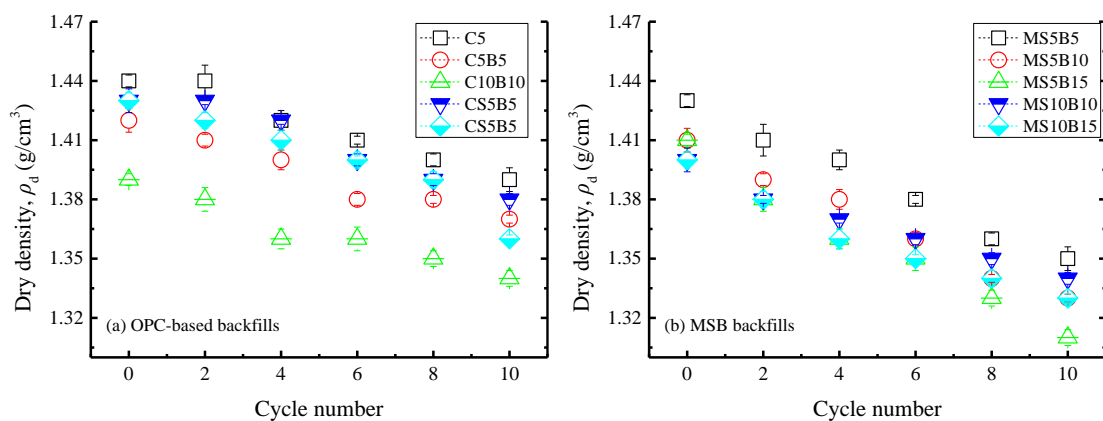
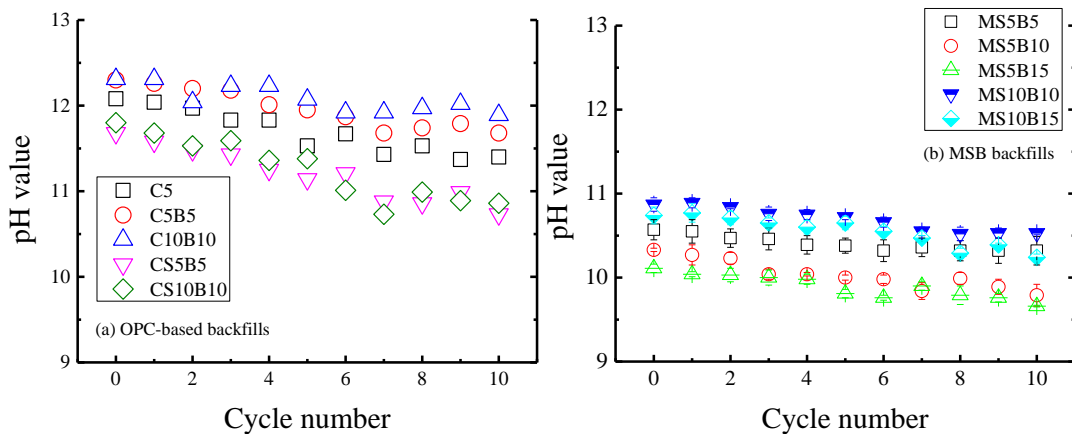


Fig 4. Dry density values for backfills after wet-dry cycles

4.3 pH value

Fig. 5 depicts the variations in the soil pH values with the wet-dry cycles for OPC-

236 based and MSB backfills. It can be observed that the pH for all backfills gradually
 237 decreases with increasing wet-dry cycles. This is attributed to the leaching of alkaline
 238 substances (i.e., Ca^{2+}) from the backfills when exposed to wetting. After ten wet-dry
 239 cycles, the pH of OPC-based and MSB backfills are 10.9 - 12.3 and 9.8 - 10.8,
 240 respectively. At the same binder and bentonite contents, the pH values of MSB backfills
 241 are much lower than those of OPC-based backfills either before or after the wet-dry
 242 cycles. For instance, the pH values for 5% GGBS-MgO in MSB backfills (i.e., MS5B5
 243 and CS5B5) are approximately 0.5 - 1.2 lower than those for 5% OPC-based backfills
 244 (i.e., C5B5 and CS5B5) after ten wet-dry cycles. Increasing the bentonite content in
 245 MSB backfills reduces the pH values by 0.4 and 0.2 unit at 5% and 10% binder contents
 246 (i.e., comparing MS5B10 and MS10B10 with MS5B15 and MS10B15, respectively)
 247 after ten wet-dry cycles. Increasing the GGBS-MgO content results in higher pH, while
 248 increasing the bentonite content decreases the pH value in the MSB mixtures.



249 **Fig. 5 Variations in pH with wet-dry cycles for (a) OPC-based and (b) MSB backfills**

250

251 **4.4 Unconfined compressive strength**

252 **Fig. 6** shows the evolution of q_u for the OPC-based and MSB specimens. It is found
 253 that q_u values of the two types of backfill decrease with increasing wet-dry cycles. After
 254

255 ten wet-dry cycles, the q_u values of the OPC-based and MSB backfills are 400 – 540
 256 kPa and 180 – 420 kPa, respectively. The decreased q_u is attributed to the gradually
 257 dissolved Ca^{2+} from the OPC-based and MSB matrices exposed to wetting conditions
 258 (Du et al., 2016; Shen et al., 2008). As the pH decreases to below 10.8 (Fig. 5), the
 259 partial destruction of C-S-H gels in the MSB backfill matrix also leads to lower q_u
 260 when compared to OPC-based backfills (Fig. 6). For the MSB backfills, the q_u of 10%
 261 GGBS-MgO in MSB backfills (i.e., MS10B10 and MS10B15) is 2.1 - 2.3 times higher
 262 than that of 5% GGBS-MgO (i.e., MS5B10 and MS5B15). It is also observed that
 263 increasing the bentonite content at the same binder content slightly reduces the strength
 264 of MSB mixtures, and that is consistent with the higher mass change and lower dry
 265 density shown in Fig. 2 and Fig. 4.

266 The strength loss (SL) and cumulative strength loss (CSL) is computed by
 267 Equations (2) and (3).

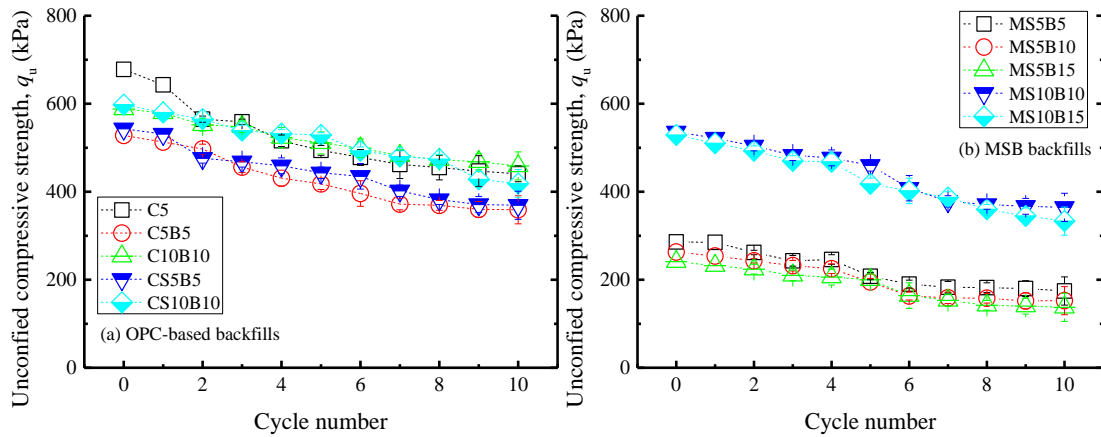
$$268 \quad SL_i = (q_{u0} - q_{ui}) / q_{u0} \quad (2)$$

$$269 \quad CSL = \sum_{i=0}^{10} SL_i \quad (3)$$

270 where q_{u0} = initial unconfined compressive strength of the specimen measured prior to
 271 the wet-dry cycles, and q_{ui} = mass unconfined compressive strength of the sample
 272 measured immediately after the i^{th} wet-dry cycle.

273 It can be observed that the CSL for 5% GGBS-MgO backfills (i.e., MS5B10 and
 274 MS5B15) is 35% - 47% higher than that of 10% GGBS-MgO (i.e., MS10B10 and
 275 MS10B15) as shown in Fig. 7. Furthermore, CSL decreases by 2.7% and 6.4% for 5%
 276 and 10% GGBS-MgO in MSB backfills, respectively, as the bentonite increases from

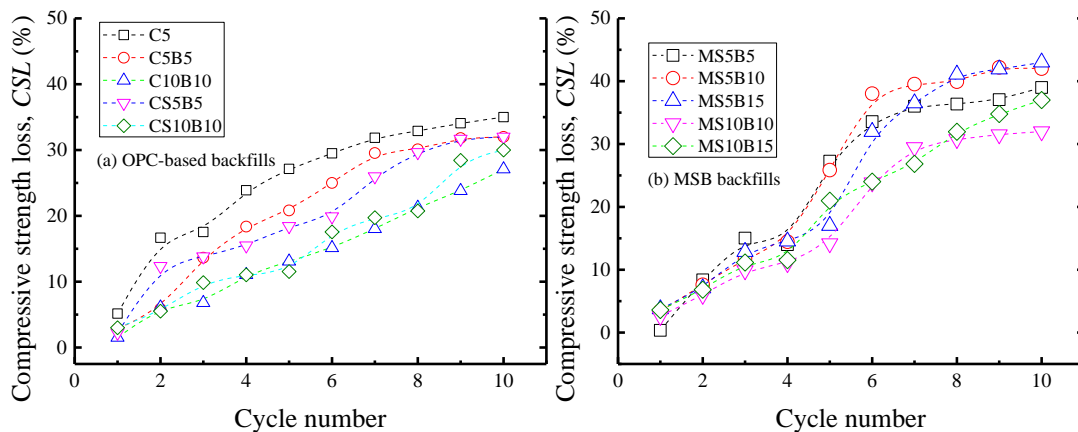
277 10% to 15%. Thus, the impact of wet-dry cycles on MSB backfills is greater than on
 278 OPC-based backfills. This phenomenon might be attributed to: (i) a lower Ca/Si ratio
 279 of C-S-H in MSB backfills as compared to that formed in the OPC-based backfills,
 280 which is more prone to shrinkage under the drying process, resulting in micro-cracking
 281 of the matrix (Jin et al., 2014b) and (ii) a larger amount of uncombined water remaining
 282 in the MSB backfills due to the better preserved bentonite swelling/shrinkage behavior
 283 (Wu et al., 2019), which is prone to evaporation and leads to substantial shrinkage
 284 cracking upon drying, resulting in greater strength loss.



285

286

Fig. 6 Effective of wet-dry cycles on q_u for (a) OPC-based and (b) MSB backfills



287

288

289

Fig. 7 Effect of wet-dry cycles on cumulative strength loss for (a) OPC-based and (b) MSB backfills

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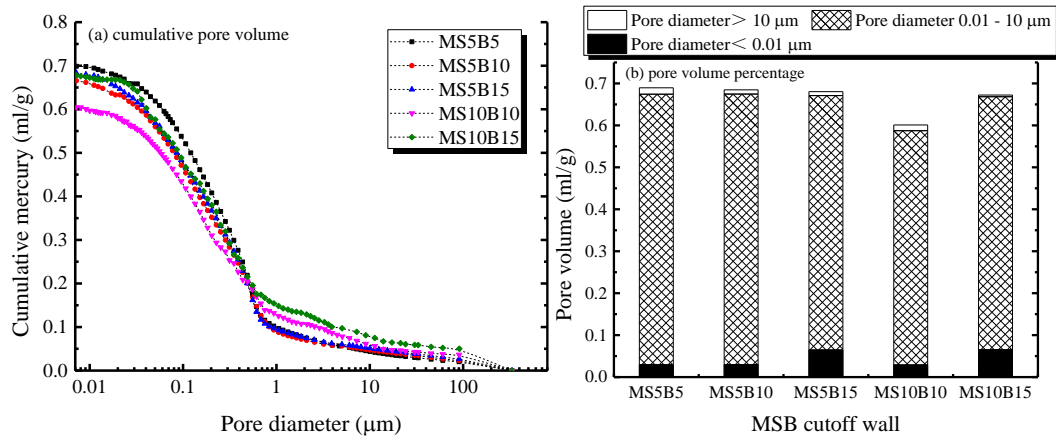
4.5 Pore size distribution

291

To investigate the evolution in pore profiles of MSB backfills with respect to wet-

292 dry cycles, MIP tests are conducted. **Fig. 8 (a)** shows the cumulative pore volumes of
293 the MSB backfills after ten wet-dry cycles. At the same bentonite content, the
294 cumulative pore volumes of MS5B10 and MS5B15 are found to decrease by 12% and
295 2% as compared with MS10B10 and MS10B15. At 5% GGBS-MgO content, increasing
296 the bentonite content from 5% to 10% and 15% increases the cumulative pore volume
297 by 4.6% and 7.9%, respectively. The results for the pore profiles are consistent with the
298 observed trends for q_u and mass change as shown in **Fig. 6 (b)** and **Fig 2 (b)**.

299 **Fig. 8 (b)** shows the volumes of the pores in different size ranges: $<0.01 \mu\text{m}$ (intra-
300 aggregate), $0.01 - 10 \mu\text{m}$ (inter-aggregate) and $>10 \mu\text{m}$ (air pores) respectively. This
301 classification of pore sizes is suggested by [Horpibulsuk et al., \(2010\)](#) and [Xia et al.,](#)
302 [\(2018 and 2019\)](#) for the OPC stabilized silty clay. Regardless of the binder content, the
303 proportions of air pores and inter-aggregate pores decrease but intra-aggregate pores
304 increase as the bentonite content increases from 5% to 15%. At the same bentonite
305 content, increasing the binder content decreases the proportions of pores in each
306 category (i.e., comparing MS5B10 and MS10B10 with MS5B15 and MS10B15). As
307 reported in previous studies ([Collins and Sanjayan, 2000](#); [Du et al., 2014 and 2016](#)), a
308 larger volume of inter-aggregate pores can lead to higher drying-initiated capillary
309 tension forces and greater shrinkage potential in the matrix. Thus, the MS10B10
310 mixture shows the highest q_u and least mass change among all the MSB backfills as
311 shown in **Fig. 6** and **Fig. 2**.



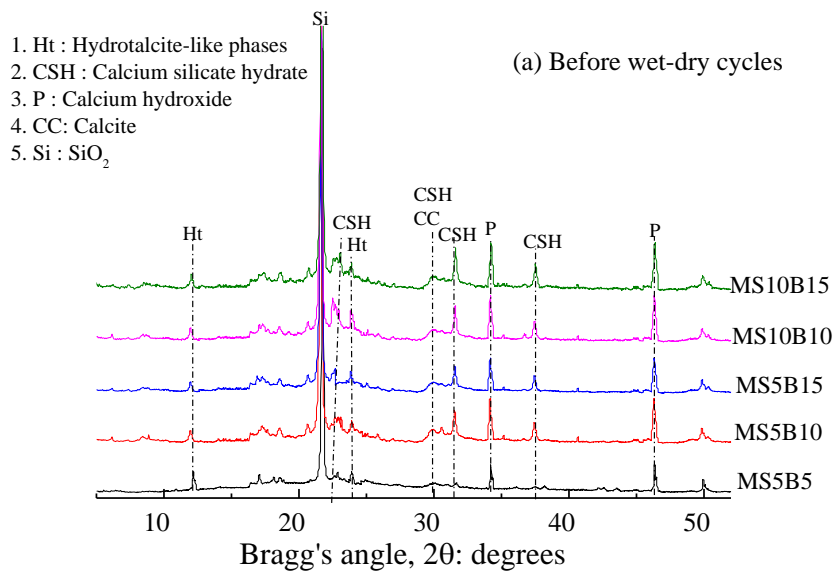
312

313 **Fig. 8 The (a) cumulative pore volume and (b) pore volume percentage of MSB backfills**
 314 **after 90 days standard curing and subjected to ten wet-dry cycles**

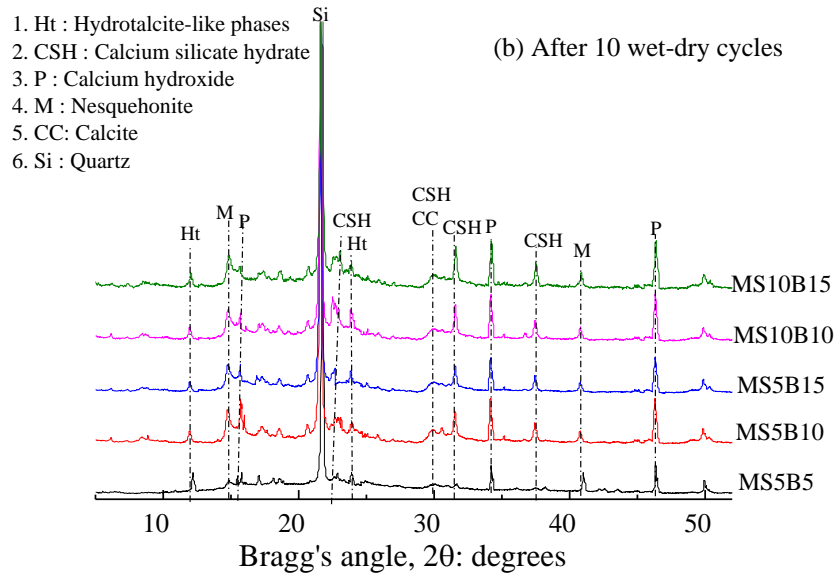
315 4.6 X-ray diffraction

316 XRD tests were conducted on the MSB specimens before and after ten wet-dry
 317 cycles to investigate the evolution of the phase assemblage in the mixtures as presented
 318 in **Fig. 9 (a) and (b)**. The characteristic peaks of hydrotalcite
 319 ($\text{Mg}_6\text{Al}_2(\text{OH})_{16}\text{CO}_3 \cdot 4\text{H}_2\text{O}$)-like phases (Ht) were found at $2\theta \approx 12.5^\circ$ and 24.0° ,
 320 agreeing well with previous findings (Jin et al., 2014a). In addition, two broad peaks
 321 ascribed to C-S-H were detected at $2\theta \approx 23.0^\circ$, 30.0° , 31.6° and 37.5° , and were the
 322 result of the reaction between released silicate species and Ca from GGBS (Wu et al.,
 323 2019; Jin et al., 2014a and b). The C-S-H gels and hydrotalcite can improve the strength
 324 and hydraulic performance in MSB cutoff wall applications. Further, hydrotalcite has
 325 contaminants (e.g., lead and zinc) with a high capacity for adsorption and which
 326 therefore can enhance the chemical compatibility of the backfills exposed to
 327 contaminated underground conditions. The higher intensity of Ht and C-S-H are
 328 observed in 10% GGBS-MgO (i.e., MS10B10 and MS10B15) as compared with 5%
 329 GGBS-MgO (i.e., MS5B5, MS5B10 and MS10B15), and result in higher q_u (Fig. 6)

330 and lower mass change (**Fig. 2**). The characteristic peak of quartz (SiO_2) has been
 331 detected at $2\theta \approx 21.6^\circ$ in the local clayey sand. The characteristic peaks of calcium
 332 hydroxide ($\text{Ca}(\text{OH})_2$) have been detected at $2\theta \approx 19.0^\circ$, 34.5° and 46.5° , from the
 333 leaching of Ca^{2+} from GGBS and C-S-H. The leaching of Ca^{2+} will lead to the decrease
 334 of pH value as illustrated in **Fig. 5**. Nesquehonite ($\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$) is found in the
 335 backfills after ten wet-dry cycles, as the peaks at $2\theta \approx 14.2^\circ$ and 41.0°) agree with those
 336 reported by [Wu et al., \(2018b\)](#) and [Ruan et al. \(2019\)](#) It is formed by the dissolved Mg^{2+}
 337 from the MSB backfills and dissolved CO_2 from the atmosphere. Similarly the leached
 338 Ca^{2+} from the GGBS and C-S-H may react with dissolved atmospheric CO_2 to form
 339 calcite. Nevertheless, the strongest calcite peak ($2\theta \approx 30.0^\circ$) overlaps with that of C-S-
 340 H, which prevents its identification via XRD in this study.



341



342
 343 **Fig. 9 X-ray diffractograms of the 90-day cured MSB backfills before (a) and after (b) wet-**
 344 **dry cycles: Ht: Hydrotalcite-like phases, CSH: Calcium silicate hydrate, P: Calcium**
 345 **hydroxide, M: Nesquehonite, CC: Calcite, Si: Quartz**

346 **5. Limitations of current study**

347 This study reveals that the MSB mixture is more sensitive to a fluctuating
 348 groundwater condition than OPC-based backfills. This might be attributed to the
 349 hydration products (C-S-H with lower a Ca/Si ratio) and microstructure (refined pores)
 350 formed in alkali-activated GGBS paste as compared to OPC-based backfills (Du et al.,
 351 2015). Nevertheless, the unconfined compressive strength values of MSB mixtures are
 352 still above the commonly adopted design limits ($q_u \geq 100$ kPa) after ten wet-dry cycles.
 353 The much lower pH values of pore water in MSB mixtures have much weaker impact
 354 on the chemical stability of montmorillonite in bentonite as compared to OPC-based
 355 backfills (Wu et al., 2019). In addition, the use of a large amount of industrial waste
 356 gives the MSB mixture better sustainability than the OPC-based backfills.

357 Admittedly, for a full understanding of the long-term durability of the MSB
 358 backfills after wet-dry cycles, the wet-dry aging could be correlated with real time for
 359 a quantitative simulation (Shen et al., 2018 and 2019). Further investigations are

360 warranted to elucidate the change of hydraulic conductivity, hydration products, and
361 microstructures of the backfills under the various extreme environmental stresses that
362 might be encountered in the field.

363 **6. Conclusions**

364 A comprehensive laboratory testing program was conducted to reveal the effects
365 of wet-dry cycles on the properties of MSB backfills and the following conclusions can
366 be drawn:

367 (1) The mass change of OPC-based and MSB backfills increased with respect to
368 the wet-dry cycles. The MSB backfills exhibited 1.1% - 2.1% higher mass change than
369 OPC-based ones after ten wet-dry cycles. Increasing the GGBS-MgO content reduced
370 9.0 - 11% mass change but increasing the bentonite content increased mass change in
371 MSB backfills. The threshold cycle numbers for spalling and macro-cracks occurring
372 in MSB backfills were found to be in the 4th- 6th cycles.

373 (2) The dry density and pH of OPC-based and MSB backfills gradually decreased
374 with increasing wet-dry cycles. For the identical binder (OPC and GGBS-MgO) and
375 bentonite contents, the mass change of MSB backfills was higher than in OPC-based
376 backfills, while the pH was much lower.

377 (3) The unconfined compressive strength of OPC-based and MSB backfills
378 decreased with increasing wet-dry cycles. The strength values of the MSB backfills
379 with 10% GGBS-MgO were 2.1 - 2.3 times higher than those with 5% GGBS-MgO,
380 while increasing the bentonite content slightly reduced the strength. The cumulative
381 strength loss of MSB backfills was more notable than that for OPC-based backfills.

382 (4) Increasing the GGBS-MgO content from 5% to 10% reduces by 2% - 12% the
383 cumulative pore volume, while increasing the bentonite content from 5% to 15%
384 increases the cumulative pore volume by 4.6% - 7.9%.

385 (5) The hydrotalcite-like phases and C-S-H were found to be the primary hydration
386 products in the MSB backfills both before and after the wet-dry cycles. The wet-dry
387 cycles accelerated the leaching of earth alkali ions, which react with the dissolved CO₂
388 from the atmosphere to form calcite (CaCO₃) and nesquehonite (MgCO₃·3H₂O) in
389 MSB mixtures.

390

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396

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