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Author

Liu, Yue, Yan, Zhuge, Chow, Christopher WK, Keegan, Alexandra, Li, Danda, Pham, Phuong Ngoc, Huang, Jianyin, Siddique, Rafat

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# Utilization of drinking water treatment sludge in concrete paving blocks: Microstructural analysis, durability and leaching properties

Yue Liu<sup>a</sup>, Yan Zhuge<sup>a,\*</sup>, Christopher W.K. Chow<sup>a,b</sup>, Alexandra Keegan<sup>c</sup>, Danda Li<sup>a</sup>, , Phuong Ngoc Pham<sup>a,d</sup>, Jianyin Huang<sup>a,b</sup>, Rafat Siddique<sup>e</sup>

<sup>a</sup> *Natural and Built Environments Research Centre, School of Natural and Built Environment, University of South Australia, Adelaide, Australia*

<sup>b</sup> *Future Industries Institute, University of South Australia, Adelaide, Australia*

<sup>c</sup> *Australian Water Quality Centre, South Australian Water Corporation, Adelaide, Australia*

<sup>d</sup> *Faculty of Bridge and Road Engineering, The University of Danang – University of Science and Technology, 54 Nguyen Luong Bang, Danang, Vietnam.*

<sup>e</sup> *Civil Engineering Department, Thapar Institute of Engineering and Technology, Patiala, India*

*\*Corresponding author. Email address: Yan.Zhuce@unisa.edu.au*

## Abstract

The management of abundant drinking water treatment sludge (DWTS) in landfill remains an important issue. The reuse of DWTS as construction material could contribute to the development of greener concrete product and to **mitigating** the detrimental environment effect from excessive production of DWTS. This paper investigates the potential of using DWTS as sand replacement in Concrete Paving Blocks (CPB). Five CPB mixtures were designed and the replacement ratios of sand by DWTS were 0%, 5%, 10%, 15%, and 20%, by weight. Properties of CPB such as compressive strength, water absorption, abrasion resistance, sulfate attack and metal leachability were determined. The results indicated that above 10% of DWTS, **the** replacement was detrimental to such properties of the CPB. Microstructure analysis proved the addition of DWTS could result in ettringite formation and the interfacial transition zone (ITZ) between the cement matrix and DWTS was **more** porous than that of sand. In addition, the **metal** leachability test of CPB demonstrated that the addition of high-copper DWTS into CPB was safe.

Keywords: Drinking water treatment sludge; Concrete; Paving blocks; Durability properties; Leaching ability; Microstructural analysis.

## 33 1. Introduction

34 Global demand for drinking water has increased dramatically due to rapid population growth  
35 and changes in lifestyle, which means water utilities need to produce more drinking water  
36 than ever before (Ahmad et al., 2016). The conventional drinking water treatment typically  
37 involves several steps, including coagulation-flocculation, sedimentation and filtration. The  
38 term ‘drinking water treatment sludge (DWTS)’ refers to all precipitates or wastes produced  
39 during drinking water treatment processes (Ahmad et al., 2016). When aluminium sulfate is  
40 used as coagulant for water purification, the acquired DWTS contains high content of  $Al_2O_3$   
41 and  $SiO_2$ . In Australia, the DWTS annual production of various water authorities could be up  
42 to 43,500 tonnes (Maiden et al., 2015). While in the UK, the annual amount of DWTS in  
43 2014 was reported around 131,000 tonnes (De Carvalho Gomes et al., 2019). Generally, it  
44 was estimated that the daily production of DWTS had exceeded 10,000 tonnes on a global  
45 scale (Babatunde and Zhao, 2007), which has likely increased in the last decade.

46 The most common method to deal with DWTS is disposal to landfill. However, landfilling  
47 solution has become more difficult and expensive due to the larger amount of DWTS  
48 generation, the limited available lands and more stringent environmental laws (Babatunde  
49 and Zhao, 2007). For instance, the cost for DWTS disposal exceeded \$6.4 million per annum  
50 in Victoria, Australia (Maiden et al., 2015) and was £ 5.5 million in the UK (Keeley et al.,  
51 2014). Thus, developing new strategies of DWTS management, such as exploring the  
52 possibility of using DWTS as a potential construction material, instead of disposal to landfill  
53 becomes an urgent need.

54 DWTS has been used as filler material to produce bricks and ceramic products, due to similar  
55 chemical compositions with clay, especially for oxides of silica, aluminium and ferric  
56 (Anderson et al., 2003; Benlalla et al., 2015; Huang et al., 2001; Ling et al., 2017; Sarabia et

57 al., 2019). However, most of studies demonstrated the increase of sludge content decreased  
58 the strength and increased water absorption of bricks (Anderson et al., 2003; Huang et al.,  
59 2001; Wolff et al., 2015) , e.g. the compressive strength dropped from 17MPa to 7MPa with  
60 increasing sludge addition from 5% to 30% at a sintering temperature of 1000<sup>0</sup>C (Benlalla et  
61 al., 2015). Only few authors claimed the addition of sludge enhanced the mechanical  
62 performance of ceramic products up to 10% sludge replacement (Kizinievich et al., 2013).

63 Apart from ceramic applications, DWTS has a potential as an alumino-siliceous substance to  
64 synthesize geopolymer (Geraldo et al., 2017; Nimwinya et al., 2016; Tang et al., 2019a). The  
65 results indicated that the addition of DWTS could delay setting time and degrade mechanical  
66 properties of geopolymer mortar (Geraldo et al., 2017; Nimwinya et al., 2016).

67 In addition, the feasibility of using DWTS as supplementary cementitious material and to  
68 produce lightweight aggregates was studied. However, the addition of sludge also  
69 deteriorated the mechanical and durability performance of concrete specimens regardless if  
70 used as a partial replacement of cementitious material or in the production of lightweight  
71 aggregates (El-Didamony et al., 2014; Hagemann et al., 2019; Huang and Wang, 2013;  
72 Huang et al., 2005).

73 More recently, several studies attempted to use oven-dried sludge at 105 <sup>0</sup>C for 24 hours into  
74 controlled low strength material (CLSM) (Fang et al., 2018; Wang et al., 2018a). This  
75 treatment method could be more environmentally friendly and energy conserving due to  
76 relatively low-temperature requirement and no additional emission of greenhouse gas. The  
77 authors reported that 12.5% sand replacement by DWTS decreased the compressive strength  
78 from 2.2 MPa to 0.6 MPa due to the fact that organic matter could form an isolation layer on  
79 the surface of calcium ions, affecting hydration reaction. Additionally, limited research was  
80 conducted to use wet DWTS to replace sand in concrete (Ramirez et al., 2017). The results  
81 indicated that the concrete mixture with 5% DWTS content could achieve 28-day

82 compressive strength of 28MPa. However, only limited information about durability  
83 performance was available. Furthermore, the micro-structure analysis and environmental  
84 impact of oven-dried DWTS in conventional concrete products has not been researched.

85 In this study, the feasibility of utilising DWTS in Concrete Paving Blocks (CPB) has been  
86 investigated. CPB can be manufactured by dry mix method with approximately zero slump of  
87 concrete mixture. The mechanical and durability properties of CPB with varying replacement  
88 ratios of sand by oven-dried DWTS were evaluated extensively. The following tests have  
89 been conducted and discussed, such as microstructure-chemical analyses, compressive  
90 strength, abrasion resistance, water absorption, sulfate attack, and toxic characteristic  
91 leaching procedure (TCLP).

## 92 2. Materials and experimental methods

### 93 2.1 Materials

94 General Purpose (GP) cement based on AS 3972 (2010) was used as cementitious materials  
95 for the production of the CPB. The aggregates used in this study were crushed stone, sand  
96 and DWTS. The size of sand, provided by Sibelco Australia, was ranging from 8.68 $\mu$ m to  
97 1.18mm with specific gravity of 2.61 and water absorption of 0.50%. The crushed stone,  
98 sourced from Kulpara Quarry, was ranging from 2.06mm to 9.00mm, with specific gravity of  
99 2.72 and water absorption of 1.17%.

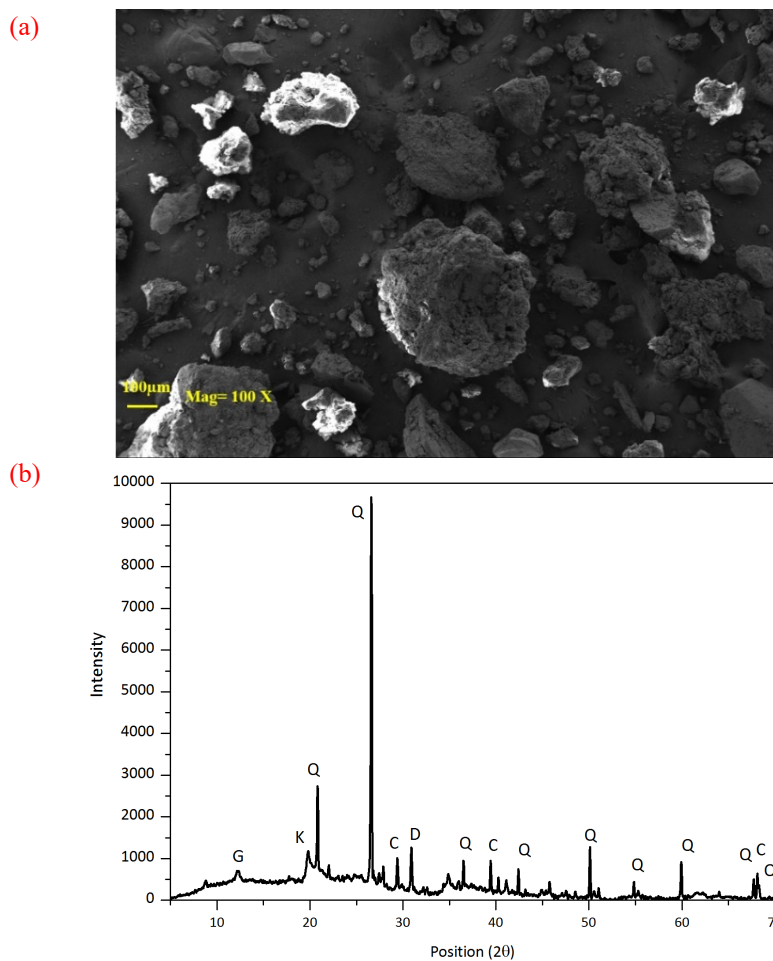
100 The DWTS was collected from Happy Valley Water Treatment Plant (Fig. S1), which is  
101 managed by SA Water and the largest water treatment facility (maximum treatment capacity:  
102 100 megalitres/day) in South Australia. The DWTS disposed to landsite on 07/2016 was  
103 chosen as research samples. The specific gravity and water absorption of DWTS were 2.27  
104 and 28.82%, respectively, based on AS 1141.5 (2000). The moisture and organic matter  
105 contents of DWTS specimen were 25% and 29.50% respectively, which were determined by  
106 loss on ignition (LOI) test according to . The chemical components of DWTS were identified  
107 by X-Ray Fluorescence (XRF) analysis (see Table 1). The results showed that Al<sub>2</sub>O<sub>3</sub> and  
108 organic matter constituted the major portion of the DWTS, followed by SiO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub> and CaO.  
109 Other oxides such as K<sub>2</sub>O, MgO, CuO, TiO<sub>2</sub> and Na<sub>2</sub>O were also observed. Referring to  
110 Scanning electron microscopy (SEM) in Fig. 1a, the morphology of DWTS particles was  
111 found to be irregular with particle sizes and porous structure. The X-ray diffraction (XRD)  
112 revealed five major crystallizations were found in DWTS, including quartz (SiO<sub>2</sub>), calcite  
113 (CaCO<sub>3</sub>), kaolinite (Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>2</sub>), gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O), and dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>)  
114 (Fig. 1b). The Toxic Characteristic Leaching Procedure (TCLP) result of raw DWTS  
115 indicated the leachability of Cu (1.8mg L<sup>-1</sup>) exceeded the limitation of USEPA (1992) (1.3mg  
116 L<sup>-1</sup>). Thus, the leachability of CPB incorporating DWTS needs to be investigated.

117 Prior to mix, DWTS was screened with No. 12 sieve (1.68mm) and the particle size  
 118 distribution of sand, crushed stone and DWTS was shown in Fig. 2, which indicated that no  
 119 noticeable difference of the particle size between sand and DWTS can be detected, and that  
 120 sand was slightly coarser than DWTS.

121 **Table 1.** Chemical compositions of DWTS.

Compositions	Weight percentage (wt%)
Al <sub>2</sub> O <sub>3</sub>	28.27
SiO <sub>2</sub>	26.43
Fe <sub>2</sub> O <sub>3</sub>	6.66
CaO	5.36
K <sub>2</sub> O	1.23
MgO	1.11
CuO	0.71
SO <sub>3</sub>	0.48
LOI	29.5

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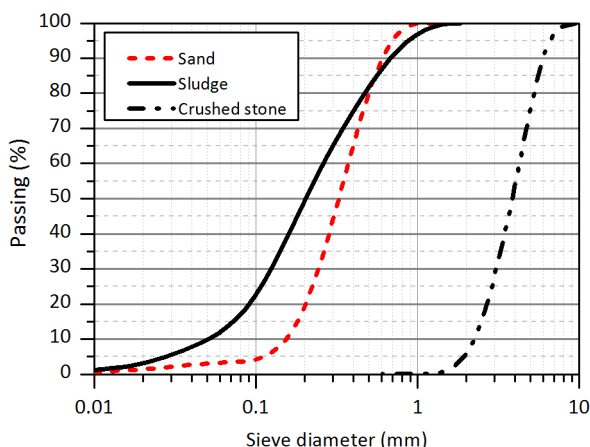


124

Fig. 1. SEM (a) and XRD spectra (b) of DWTS

125  
126

(K: Kaolinite; Q: Quartz; G: Gypsum; D: Dolomite; C: Calcite).



127  
128

Fig. 2. Particle size distribution of sand, DWTS and crushed stone.

### 129 2.2 Mix proportion of CPB

130 A total of five mixes were prepared in the laboratory scale. Based on preliminary trials, the  
 131 aggregate-to-cement ratio of 6.6 was used. In addition, the weight ratio of crushed stone to  
 132 sand was kept at 1:1.3. The water/cement ratio was fixed at 0.3 for reference mix (CPB-0).  
 133 Owing to the higher water absorption of DWTS, such ratios varied slightly with other mixes  
 134 depending on different DWTS contents in CPB, namely 5%, 10%, 15%, and 20%  
 135 corresponding to CPB-5, CPB-10, CPB-15, and CPB-20, respectively. Mix-proportion of  
 136 CPB was provided in Table 2.

137 **Table 2.** Mix proportions of CPB with varying contents of DWTS (kg/m<sup>3</sup>).

Mix notation	GP Cement	Concrete Sand	Crushed Stone	DWTS
CPB-0	299	851	1134	0
CPB-5	299	811	1134	42
CPB-10	299	767	1134	85
CPB-15	299	723	1134	129
CPB-20	299	682	1134	169

138



## 139 2.3 Manufacturing of CPB

140 CPB were produced by dry-mix method with the size of  $115 \times 115 \times 50 \text{ mm}^3$  and aggregates  
141 (crushed stone, sand and DWTS) were oven dried at  $105 \pm 3^\circ\text{C}$  for 24h before mix. For the  
142 mix, all dried ingredients were weighted and homogeneously mixed together for 1 minute.  
143 Water were then added to the mixtures before a further 3-minute mixing period. Load was  
144 applied slowly by a hydraulic compression machine and the pressure was fixed at 18MPa for  
145 30s based on general industry practice. After carefully demoulding, all samples were covered  
146 by cling wrap and stored in a chamber at a constant temperature of  $23 \pm 2^\circ\text{C}$  and relative  
147 humidity of 65% over 28-days curing period prior to carrying out the experimental testing . It  
148 should be noted that at least three replicates per mixture were produced for the following tests,  
149 except for leaching test where two samples were measured.

150

## 151 2.4 Test methods

### 152 2.4.1 Microstructural-chemical analyses

153 For SEM-based microstructural analysis, small pieces of crushed CPB taken from 2 to 4mm  
154 sample depth were oven-dried at  $40^\circ\text{C}$  for 48h and then carbon-coated. In order to detect  
155 sludge, sand and other crystallizations, Energy Dispersive X-ray spectroscopy (EDS) helped  
156 to identify chemical elements. The crystalline phases of crushed CPB samples were  
157 determined by X-ray Diffractometry (XRD). A representative pieces of CPB obtained from  
158 similar position of the samples used for SEM analysis were ground to minus  $30\mu\text{m}$ . XRD was  
159 conducted using  $\text{CuK}\alpha$  radiation at 40kV and 40mA. The scan angle was between  $5^\circ\text{C}$  and  
160  $70^\circ\text{C}$  at the scan speed of  $0.2^\circ/\text{min}$ .

161 2.4.2 Compressive test

162 Compressive strength of CPB was determined in accordance with AS 4456.4 (2003). The  
163 specimen was compressed by the universal testing machine with the wearing face up. The  
164 load was applied at a constant rate of 0.150MPa/s, without any shock, until failure.

165 2.4.3 Abrasion resistance test

166 The abrasion resistance test was set up in accordance to AS 4456.9 (2003). It involved  
167 securing the specimens to a tumbling machine with the inward faces of the specimens placed  
168 over holes on the outside of the tumbling drum. Six hundred steel balls were placed within  
169 the drum and the tumbling machine ran for 3600 revolutions at a speed of 60 rotations per  
170 minute. After the test, the samples were immersed in potable water at least 2 hours and then  
171 weighed in water and in saturated surface dry condition to get the bulk density of samples. To  
172 eliminate the atmosphere effect for this test, two samples were selected randomly from 18  
173 CPB samples as control specimens. The mass loss and abrasion index were calculated  
174 according to Eq. 1-3, respectively.

175 Mass loss (%) =  $\frac{m_1 - m_2}{m_1} \times 100$  (1)

176 Abrasion index =  $\frac{m_1 - (m_2 - C)}{B_d} \times 10^3$  (2)

177  $B_d = \text{bulk density} = \left( \frac{m_2}{m_3 - m_4} \right) \times \rho$  and  $C = m_5 - m_6$  (3)

178 Where,  $m_1$  is the initial weight of test sample (g);  $m_2$  is the weight of test sample after test  
179 (g);  $m_3$  and  $m_4$  are the weight of specimens after immersing in water and under water,  
180 respectively (g);  $m_5$  and  $m_6$  are the weight of control samples before and after test (g); C is  
181 the correction (g);  $B_d$  is the bulk density (kg/m<sup>3</sup>);  $\rho$  is the density of water (kg/m<sup>3</sup>).

#### 182 2.4.4 Water absorption test

183 According to AS 4456.14 (2003), CPB samples were oven dried at  $110\pm 8$  °C until a constant  
184 weight, before fully immersing in cold potable water for 24 h. The saturated CPB specimens  
185 were then dried with a damp cloth to remove surface water and finally weighted. The water  
186 absorption of CPB was calculated by Eq. 4, where  $m_1$  is the mass of dried sample and  $m_2$  is  
187 the mass of immersed surface dried sample (g).

$$188 \quad \text{Water absorption (\%)} = \frac{(m_2 - m_1)}{m_1} \times 100 \quad (4)$$

#### 189 2.4.5 Sodium sulfate attack test

190 There are some measurement methods to assess the resistivity of concrete products to sodium  
191 sulfate attack, such as weight loss, ultrasonic pulse velocity (UPV), compressive strength,  
192 expansion, damage variable, and visual appearance (Jiang and Niu, 2016; Nehdi et al., 2014;  
193 Pham et al., 2019; Scherer, 2004; Sotiriadis et al., 2012; Tang et al., 2019b; Yu et al., 2013).  
194 In this study, visual appearance, weight loss, and UPV of CPB were studied as the indicators  
195 of resistance of CPB to sodium sulfate attack.

196 The resistance to sulfate attack test of CPB samples was carried out according to AS 4456.10  
197 (2003). The sulfate solution was prepared by dissolving 62 g  $\text{Na}_2\text{SO}_4$  in 1L of distilled water.  
198 Each cycle of this test started by immersing the specimens in sulfate solution for 2 h and then  
199 oven dried at  $65\pm 3$  °C for  $20\pm 1$  h. During the test, pH value of solidum sulfate solution was  
200 maintained between 6-8. The mass and UPV of CPB samples were measured after cooling  
201 the specimens to room temperature ( $2\pm 0.5$  h). The test was repeated with above steps for 17  
202 cycles. The mass loss (%) was determined by Eq. 5, where  $m_0$  and  $m_i$  are weights before  
203 starting test and after  $i^{\text{th}}$  cycle of sulfate attack (g), respectively. The UPV was measured  
204 based on [ASTM C597 \(2016\)](#) and the frequency of generated ultrasonic pules was 54kHz.

205 UPV (m/s) is calculated by Eq. 6, where L is path length of pulse travel (m) and t is the  
206 measured time (s).

$$207 \quad \text{Mass loss (\%)} = \frac{m_0 - m_i}{m_0} \times 100 \quad (5)$$

$$208 \quad \text{UPV (m/s)} = \frac{L}{t} \quad (6)$$

#### 209 2.4.6 Toxic characteristic leaching procedure (TCLP)

210 Due to the hazardous leaching problem of raw DWTS discussed in section 2.1, TCLP was  
211 conducted for CPB. The leachability of metal elements was the main concern for applications  
212 of DWTS in CPB. According to the USEPA (1992), the crushed samples were taken from the  
213 specimens after compressive strength tests and were **screened** by a 10mm sieve. For each  
214 CPB, 20 g of crushed sample was put into 400mL of the TCLP leachate (5.7mL of glacial  
215 acetic acid in 2 L of distilled water) and then shaken for 18 h on an automatic shaker (Ling  
216 and Poon, 2014). The concentration of different metals in the leachate was determined by  
217 Agilent 8800 Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

218

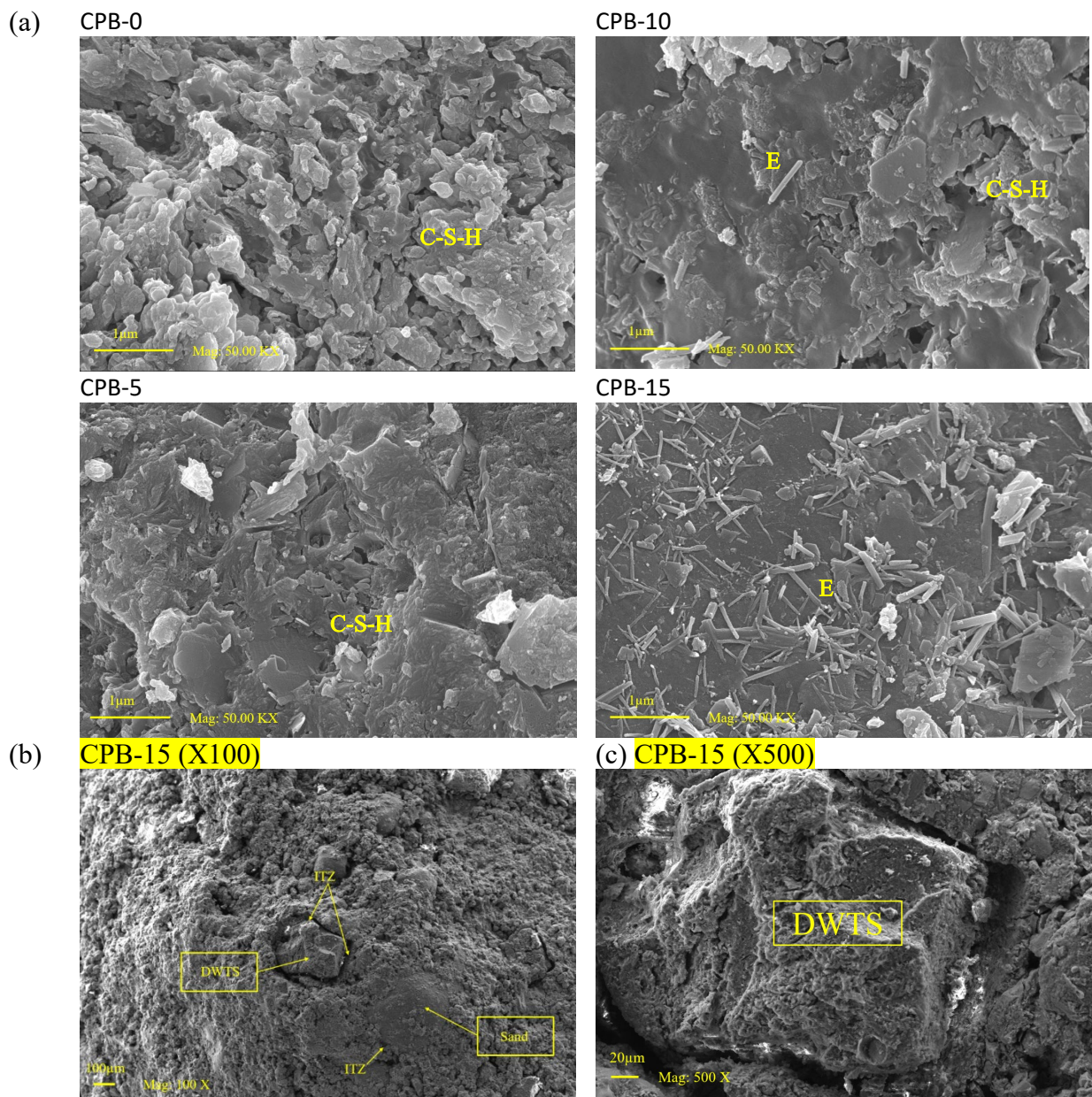
### 219 3. Results and discussion

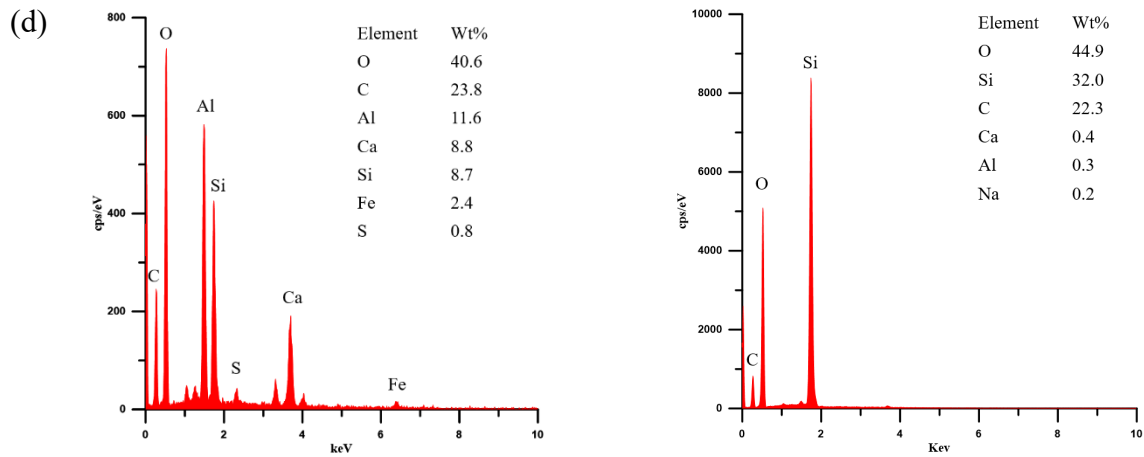
#### 220 3.1. Microstructural chemical characterizations

##### 221 3.1.1. Microstructural observations

222 Fig. 3a shows the SEM images of CPB with DWTS content varying from 0% to 15%. The  
223 dominated compound in CPB-0 was the cotton-shaped C-S-H (calcium silicate hydrate) gel,  
224 which was the main hydration product of Portland cement. A similar microstructure to CPB-0  
225 was also observed on CPB-5. For CPB-10, the C-S-H gel was not evenly distributed  
226 compared with CPB-0. Needle-shaped ettringite was also revealed on CPB-10 and especially  
227 more visible on CPB-15. **However, the cotton-shaped C-S-H gel was not visible in CPB-15.**

228 On one hand, the difference in SEM images of CPB samples with higher contents of DWTS  
 229 could be due to the organic matter in DWTS which hindered the formation of portlandite and  
 230 C-S-H gel (Wang et al., 2018b). On the other hand, it could be due to the chemical  
 231 composition of DWTS, which contained alum and gypsum. Initially, the  $\text{Ca}^{2+}$  ions from  
 232 portlandite or gypsum dissolved in solution reacted with  $\text{C}_3\text{A}$  and sulfate ions to form  
 233 ettringite. Furthermore, the sulfates reacted with the monosulfate and  $\text{Ca}^{2+}$  ions to produce  
 234 more ettringite, where  $\text{Ca}^{2+}$  was provided by CH or C-S-H after depletion of CH (Menéndez  
 235 et al., 2013).





236 Fig. 3. SEM-EDS analysis of CPB (a) SEM images of CPB with varying contents of DWTS (CSH: Calcium-  
 237 Silicate-Hydrate; E: Ettringite), (b) ITZ between aggregates (sand and DWTS) and cement matrix (100X) in  
 238 CPB-15, (c) ITZ between DWTS and cement matrix (500X) in CPB-15, (d) EDS analysis for DWTS and sand.

239

240 It is well known that the strength of concrete is affected by strength of cement matrix,  
 241 aggregates, and the interfacial transition zone (ITZ) between cement matrix and aggregates.

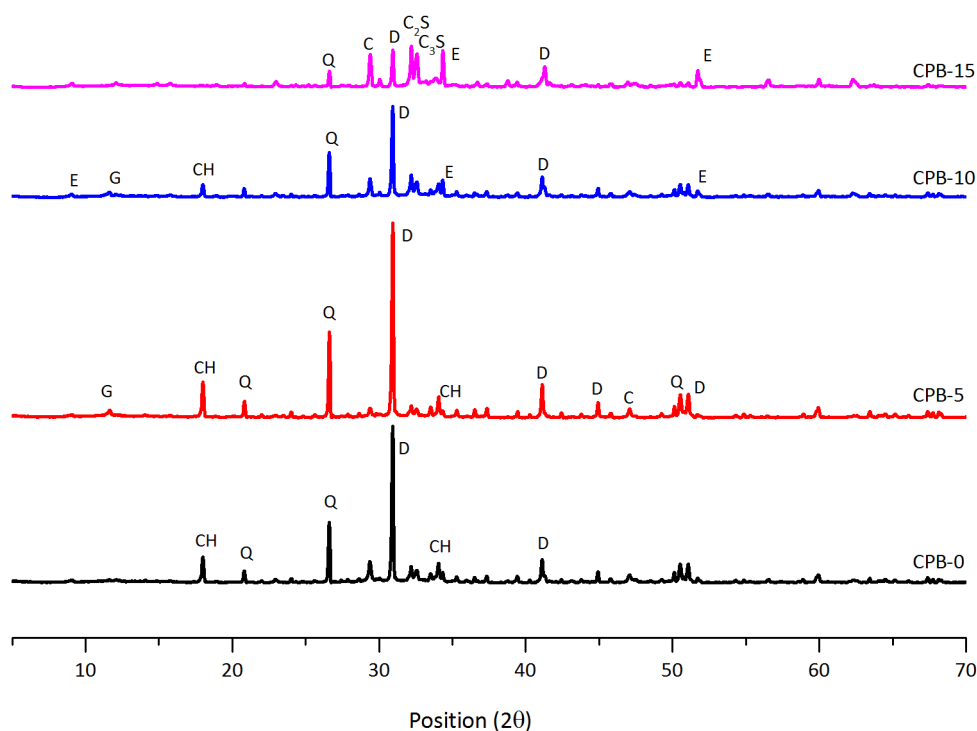
242 ITZ has been recognized as the weakest phase in concrete and the denser and smaller  
 243 thickness of ITZ indicate a stronger interfacial bonding between aggregate and cement matrix  
 244 (Poon et al., 2004). Fig. 3b and 3c shows the interfaces of cement-DWTS and cement-sand in

245 CPB samples. Fig. 3d identified the substance of corresponding areas by EDS, which showed  
 246 DWTS and sand, respectively. Generally, the DWTS-cement interfaces observed from SEM  
 247 images were very loose, and the thickness of ITZ was varying along the surface of DWTS,  
 248 approximately between 30-40 $\mu$ m (Fig. 3b). Fig. 3c shows a higher magnification of DWTS-  
 249 cement interface, it exhibits the ITZ is porous and some loose particles stockpiles in this area.

250 It can be explained by the high-water absorption capacity of DWTS, which absorbs a large  
 251 amount of water, resulting in lack of free water for cement hydration. Thus, the wider and  
 252 more porous ITZ was observed in hardened CPB incorporating DWTS. Compared with  
 253 DWTS, the sand-cement interface was relatively denser with maximum thickness around  
 254 15 $\mu$ m (see Fig. 3b).

255 3.1.2 Composition analysis

256 Fig. 4 shows the XRD spectra of CPB with 0%, 5%, 10% and 15% DWTS contents. For the  
257 reference specimens (CPB-0), obvious diffraction peaks for CH, and quartz and dolomite  
258 could be observed. CPB-5 exhibited similar diffraction pattern to CPB-0, except for the  
259 presence of gypsum, which could be derived from DWTS. Compared with CPB-5 and CPB-0,  
260 CH proportion was significantly reduced and some diffraction peaks of ettringite were  
261 observed in the CPB-10. Furthermore, CH peaks were noticeably diminished in CPB-15.  
262 Instead, a significant increase in the peak intensity of CPB-15 at 32.3<sup>o</sup>C associated with two  
263 overlapping peaks of C<sub>2</sub>S and C<sub>3</sub>S were observed. It confirmed the addition of DWTS may  
264 hinder the cement hydration. In addition, the diffraction peaks of ettringite in CPB-15 was  
265 higher than that of CPB-10, which proved large content of DWTS could result in formation  
266 of ettringite. The peak of gypsum in CPB-15 was not as clear as CPB-10 and CPB-5,  
267 attributing to the formation of ettringite consumed gypsum.



268

269 Fig. 4. XRD spectra of CPB with varying contents of DWTS (Q: Quartz; C: Calcite; E: Ettringite; D: Dolomite;  
270 G: Gypsum; CH: Portlandite; C<sub>2</sub>S: Dicalcium silicate; C<sub>3</sub>S: Tricalcium silicate).

271

## 272 3.2 Mechanical and durability properties

### 273 3.2.1 Compressive strength

274 **Table 3** shows the compressive strength of five mixes at 28-days of curing. The **compressive**  
275 **strengths of** CPB with 0%, 5%, 10%, 15% and 20% contents of DWTS were 48.69MPa,  
276 **52.20MPa**, 43.25MPa, 9.92MPa and 4.13MPa, respectively. **The compressive strength of**  
277 **CPB-5 was comparable with the control mix** (CPB-0). However, compared to CPB-0, the  
278 compressive strengths of CPB-10, CPB-15, and CPB-20 were lower, namely 11.2%, 79.6%,  
279 and 91.5%, respectively. The strength requirement of CPB in Australia was 43MPa (**Ghafoori**  
280 **and Smith, 1992**). Thus, the obtained results indicated the maximum replacement ratio of  
281 sand by DWTS in CPB was 10%.

282 **The adverse effect of incorporating DWTS on compressive strength can be attributed to the**  
283 **loss of cohesion between DWTS and cement matrix as observed in Fig 3 and hindering of**  
284 **cement hydration. The DWTS was characterised with high-water absorption rate and high**  
285 **content of organic matter. The increased DWTS content in CPB could result in lack of water**  
286 **for cement hydration**, since a larger amount of free water was absorbed by oven-dried DWTS.  
287 In addition, the organic matter contained in DWTS (29.5%) hindered the formation of  
288 hydration **products** (Wang et al., 2018a). Thus, **the deterioration of CPB** was getting more  
289 severe with the increasing of DWTS replacement levels. Eventually, the strength  
290 development of CPB with 20% untreated DWTS content was almost invisible.

291

### 292 3.2.2 Abrasion resistance

293 Due to completed disintegration of CPB-15 and CPB-20 within 15 minutes from the start of  
294 the experiment, the test results only include CPB-0, CPB-5, and CPB-10. The effect of  
295 addition of DWTS on abrasion resistance of CPB is shown in Table 3. The average abrasion



index for CPB-0, CPB-5, and CPB-10 were 11.91, 12.23, 12.42, respectively. The results confirmed that the addition of DWTS up to 10% had no significant effect on the resistance of CPB to abrasion, which can be related to the relatively good strength of CPB-0, CPB-5 and CPB-10 and good compatibility of DWTS in CPB under 10% replacement level.

**Table 3. Strength and durability properties of CPB with varying contents of DWTS.**

Mix notation	Compressive strength (MPa)	Abrasion properties			Water absorption (%)
		Mass loss (%)	Abrasion Index	Notes	
CPB-0	48.69(±2.3)	1.9(±0.1)	11.91(±0.32)		4.2(±0.35)
CPB-5	52.20(±1.9)	1.6(±0.2)	12.23(±0.52)		3.8(±0.15)
CPB-10	43.25(±2.2)	2.1(±0.2)	12.42(±0.56)		5.1(±0.25)
CPB-15	9.92(±1.4)	-	-	Specimens failed	7.3(±0.20)
CPB-20	4.13(±0.8)	-	-	Specimens failed	7.7(±0.15)

301

### 302 3.2.3 Water absorption

303 The water absorption of the five DWTS contents at 28-days are shown in Table 3. It can be  
 304 seen that the water absorption of CPB with 0%, 5%, 10%, 15%, and 20% DWTS replacement  
 305 were 4.2%, 3.8%, 5.1%, 7.3% and 7.7%, respectively. The CPB-0, CPB-5 and CPB-10  
 306 demonstrated relative low water-absorption. The CPB with 15% and 20% DWTS  
 307 replacements did not satisfy the criteria for water absorption (less than 6%) (Jankovic et al.,  
 308 2012). Generally, the water absorption of CPB increased with the addition of DWTS. This  
 309 can be explained by the porous structure of CPB incorporating DWTS which resulted in  
 310 higher water absorption. However, the CPB with 5% DWTS content demonstrated a denser  
 311 structure, which resulted in lower water-absorption ability.

### 312 3.2.4 Sulfate attack

#### 313 3.2.4.1 Mass loss

314 Fig. 5a shows the mass loss of CPB with the five DWTS contents. The increase in mass for  
 315 CPB with DWTS contents of 0%, 5%, and 10% were 3.2%, 3.8%, and 3.2% after 17 cycles

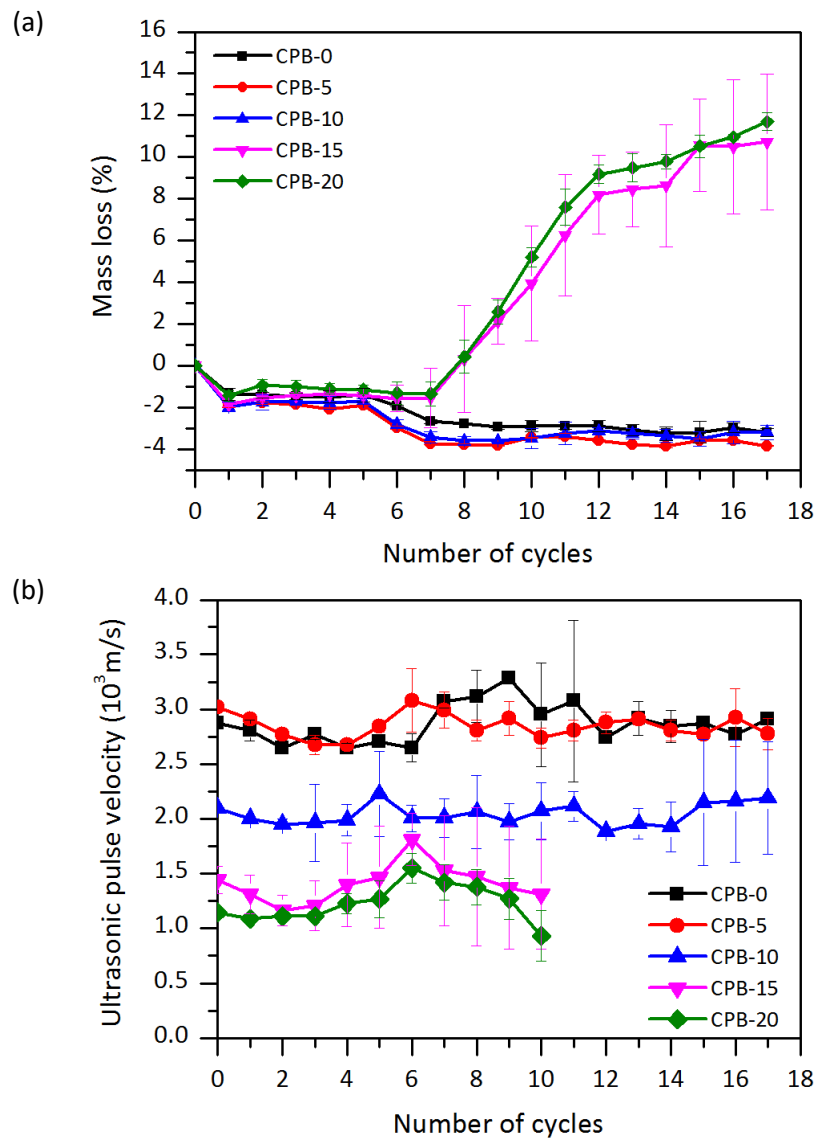
316 of salt attack, respectively. The mass of CPB-15 and CPB-20 both started to decrease at the  
317 7<sup>th</sup> cycle, and the final mass losses were 10.72% and 11.69%, respectively. Furthermore, the  
318 reduction rate in the mass of CPB-20 was slightly higher than CPB-15.

319 Due to wetting and drying cycles in **experimental** process, **the deterioration of CPB samples**  
320 **could be from a combination of physical and chemical attack.** For instance, the sodium  
321 sulfate solution could be absorbed by capillary suction and the subsequent evaporation of  
322 water could contribute to the deposit of salt in pores close to surface of CPB (Scherer,  
323 2004).**The chemical attack was characterized by chemical reactions between sulfate and**  
324 **aluminate components in hardened cement pastes to produce expansive ettringite and gypsum,**  
325 **leading to loss of concrete pieces (Bakhareva et al., 2002).**

326 The increase in weight associated with CPB-0, CPB-5 and CPB-10 could be attributed to the  
327 gradual infiltration of sulfate solution into the capillary voids of CPB and the consequent  
328 reaction with alumina-containing hydrates forms ettringite. The formed ettringite can fill the  
329 voids of the composite matrix and result in the increase of mass. For the significant weight  
330 loss of CPB-15 and CPB-20, it can be explained by the porous structure of DWTS and the  
331 high contents of DWTS. Compared with CPB-0, CPB-5, and CPB-10, the pavers  
332 incorporating higher sludge contents (15% and 20%) with more porous microstructure were  
333 easier to be penetrated by the salt solution and higher aluminium content was detrimental to  
334 sulfate resistance (Bakhareva et al., 2002). The excessive formation of ettringite caused  
335 tensile stresses to develop, leading to crack initiation and CPB deterioration when such  
336 stresses became greater than **the** tensile strength of the composites (Pham et al., 2019). In  
337 addition, **the** accumulation of salt crystallization during the wetting cycle could cause spalling  
338 and cracking due to the thenardite reprecipitating as mirabilite (Scherer, 2004).

339 Fig. 6 presents the images of typical specimens with DWTS content from 0 to 20% after 17  
 340 cycles of salt attack. Minor spalling of surface skin was evident on the corner and edges of  
 341 CPB-0, CPB-5 and CPB-10. For CPB-15 and CPB-20, the surface spalling was much more  
 342 serious than CPB-0, CPB-5 and CPB-10. No cracking was noticed in all mixtures. Only CPB-  
 343 0, CPB-5, and CPB-10 can meet the requirements of AS 4455.2 (2010), which specified the  
 344 maximum weight loss should not exceed 0.4g after 17 cycles of the experiment.

345  
 346



347 Fig. 5. Mass loss (a) and change of ultrasonic pulse velocity (b) with varying replacement ratios of DWTS.

348  
 349

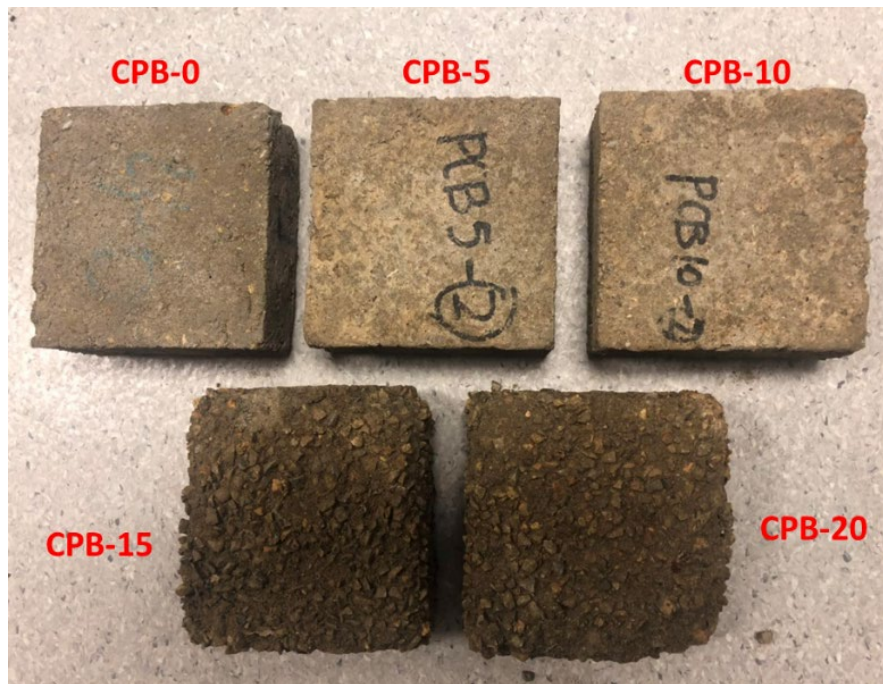


Fig. 6. Degradation of CPB samples after 17 cycles of salt attack.

350

351

352

#### 353 3.2.4.2 Ultrasonic pulse velocity (UPV)

354 UPV is a non-destructive test method, which can detect the interior of the specimen and  
355 provide evidence of the micro-crack inside concrete block (Tang et al., 2019b). The UPV of

356 CPB over 17 cycles of sulfate attack is shown in Fig. 5b. Note that, due to severe  
357 deterioration of CPB surface exposed to sulfate attack, the velocity measurement for CPB-15

358 and CPB-20 were only carried out for the first ten cycles. The initial UPV of CPB-5 was

359 quite identical to CPB-0. This was attributed to the similar dense micro-structure which  
360 reduced transmitting time of ultrasonic pulse waves, leading to the higher pulse velocity.

361 Under the condition of sodium sulfate attack, pulse velocities of CPB-0, CPB-5, and CPB-10  
362 had no significant change compared with their original values. On the other hand, CPB-15

363 and CPB-20 exhibited a higher rate of velocity increase at 6<sup>th</sup> cycle due to the formation of  
364 ettringite, which helped to densify the microstructure of composite. However, the extreme

365 development of the expansive product resulted in cracks and consequently the UPV was  
366 significantly decreased until fractured.

367

368 3.3 Toxic characteristic leaching procedure

369 Table 4 reports the leaching behaviour of raw DWTS and CPB with varying DWTS contents.

370 The results indicated that the leaching concentrations of the listed elements of CPB with  
371 varying DWTS contents were low, and all the detected heavy metals were within the  
372 limitation based on (USEPA, 1992).

373 The leaching rate of the CPB was mainly controlled by capillary diffusion. During the  
374 immersing process, water penetrated into the CPB due to the existence of capillary voids,  
375 therefore chemical elements can dissolve into water. When water attained the potential  
376 maximum depth gradually, the elemental leaching rate was decreased significantly, and the  
377 leaching concentrations of elements appeared to be stable. Thus, the leaching concentrations  
378 of Al and Cu generally increased with the increase of DWTS contents from 5% to 20% in  
379 CPB, which allowed water to be transported quicker due to the higher porosity of the  
380 composites. The obtained results indicated that the cement-based material was efficient to  
381 immobilise metal elements and the addition of DWTS into CPB was safe.

382 **Table 4.** Leachate concentration of DWTS and CPB samples (mg L<sup>-1</sup>).

Samples	Al	Cr	Mn	Ni	Cu	Zn	As	Ba	Pb	Cd
DWTS	51.504	0.003	4.222	0.022	1.810	0.063	0.006	0.580	0.002	0.001
CPB-5	0.057	0.035	0.520	0.024	0.020	0.062	0.002	0.196	0	0
CPB-10	0.121	0.042	0.334	0.021	0.030	0.032	0.001	0.032	0	0
CPB-15	0.944	0.046	0.776	0.024	0.081	0.063	0.001	0.063	0.002	0
CPB-20	2.136	0.046	0.494	0.019	0.125	0.080	0.002	0.080	0.002	0.001
Regulatory Limits	-	0.6	-	-	1.3	4.3	10.2	-	0.75	1.53

383

## 384 4. Conclusions

385 This study aimed to explore the potential possibility of adding DWTS into CPB as a  
386 replacement for sand. The experimental results proved that DWTS can be used to replace fine  
387 aggregates and the maximum DWTS contents in CPB could be up to 10% depending on  
388 paving criteria. From the analysis of experimental results, some conclusions can be drawn:

- 389 (1) The microstructural analysis revealed high porosity of CPB with DWTS  
390 incorporation, especially at the ITZ between DWTS and cement matrix. Ettringite  
391 was also formed in the composites incorporating high DWTS content due to sulfur  
392 and alum available in DWTS.
- 393 (2) CPB exhibited a reduction in 28-day compressive strength with increasing of  
394 DWTS content, except for CPB with 5% DWTS. The strength decreased  
395 significantly when higher DWTS contents were used (15% and 20%) due to a  
396 more porous structure and higher organic matter in DWTS.
- 397 (3) The increasing content of DWTS was detrimental to abrasion resistance and  
398 increased water absorption of CPB. CPB with above 10% of DWTS did not  
399 satisfy the requirement for water absorption of pavers and also was damaged  
400 quickly after starting the abrasion test.
- 401 (4) For the durability of pavers under aggressive environments, CPB incorporating up  
402 to 10% of DWTS could maintain the resistance of the composites to sodium  
403 sulfate attack as reference mix (CPB-0). Severe spalling was observed on CPB  
404 with higher DWTS content due to both chemical and physical sodium sulfate  
405 attack.
- 406 (5) In spite of the high leachability of Cu in DWTS used, this element and all the  
407 other metals in DWTS were well immobilised in CPB, so the obtained product  
408 was non-hazardous for paving application.

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