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5

6 **Studying the Mix Design and Investigating the Photocatalytic Performance of Pervious**
7 **Concrete Containing TiO₂-Soaked Recycled Aggregates**

- 8 • The research used recycled aggregates (RAs) in pervious concrete for air purification
9 purpose.
- 10 • RAs could absorb more nano photocatalysts.
- 11 • The study identified the optimized mix design to achieve highest pervious concrete
12 strength.
- 13 • The optimized concentration of TiO₂ solution was identified at 0.3% in order to achieve
14 the highest NO degradation rate at 70% for pervious concrete.
- 15 • Previous concrete showed a more durable photocatalytic performance after rain wash.
16

17 **Abstract**

18 Demolished concrete, as one main form of construction and demolition wastes, has been widely
19 studied of being utilized as recycled aggregates (RAs) in new concrete production. However,
20 existing studies of applying RAs have been limited to the mechanical and durability issues of
21 cementitious composites containing RAs. There has not been sufficient research of adopting RAs
22 in cementitious products to also address the environmental sustainability. On the other hand,
23 existing research utilizing cementitious products (e.g., concrete pavement) for air purification
24 purpose have not adequately considered RA usage. Aiming to address the two sustainable
25 objectives (i.e., waste diversion and air purification) simultaneously in concrete mix, this
26 research adopted a two-step approach. Firstly, we studied and identified the optimal mix design
27 of pervious concrete containing TiO₂-soaked recycled coarse aggregates (RCAs) in order to
28 achieve the higher compressive strength; secondly, we investigated the photocatalytic

29 performance of pervious concrete containing RCAs coated with TiO_2 photocatalysts. The
30 photocatalytic performance of pervious concrete was also tested by applying a 10-min heavy
31 rainwater wash. Experimental test results revealed that the internal voids of adhered mortar
32 enabled RCAs to absorb more TiO_2 particles. The NO degradation rate of TiO_2 -soaked RCAs
33 increased from 71.4% to 80.6% when RCAs' size decreased from 15-20 mm to 5-10 mm. The
34 orthogonal experimental investigation indicated that water-to-binder ratio had the most
35 significant effect on concrete compressive strength, followed by ratio of RCAs to binder, and
36 replacement ratio of RCAs to natural aggregates. The optimized mix design for pervious
37 concrete containing RCAs was identified to achieve highest strength (i.e., water-to-binder ratio at
38 0.35, coarse aggregate-to-binder ratio of 3 by mass, fly ash replacement rate to Portland cement
39 at 5%, and 50% replacement ratio of RCAs to NCAs,). The concentration of TiO_2 solution at
40 0.3% was identified as the optimal ratio to achieve the highest NO degradation rate at 70%
41 before rainwater wash. The NO degradation rates of pervious concrete still reached nearly 50%
42 after 10-min heavy rainwater wash, indicating that pervious concrete using RCAs coated with
43 TiO_2 could largely maintain its photocatalytic capacity. This study addresses two main social and
44 environmental issues in developing countries (e.g., China), namely overwhelming amount of
45 construction & demolition wastes being generated, and air pollution. It leads to the cleaner
46 production in concrete pavement construction by achieving the optimization between waste reuse,
47 air purification, and engineering properties of porous concrete.

48 **Keywords:** Pervious concrete; recycled aggregate concrete; photocatalytic effects; mechanical
49 properties; recycled aggregate; titanium dioxide

50 **1. Introduction**

51 Concrete is the most widely consumed building material, and the production of concrete is

52 causing wide sustainability concern due to its consumption of natural resources (Mobasher,
53 2008). Concrete itself accounts for 50% to 70% of construction and demolition (C&D) wastes
54 worldwide (Kim and Kim, 2007; Tam, 2008). Developing countries such as China is generating
55 more C&D wastes due to its infrastructure development and urbanization (Jin *et al.*, 2018a; Shi
56 *et al.*, 2016). The urgency of reducing the landfill demand in developing economies like China
57 asks the sustainable treatment of C&D wastes (Jin *et al.*, 2017). The possibility of recycling
58 construction wastes, particularly concrete, has become a major issue worldwide (Xiao *et al.*,
59 2015). Recycling old concrete as aggregates for new concrete production (i.e., recycled
60 aggregate concrete) is one of the effective approaches to achieve sustainable concrete (Xiao *et al.*,
61 2015). Numerous studies have investigated how the recycled aggregate (RA) affected the
62 properties of concrete containing recycled ingredients (e.g., RA), including mechanical
63 properties (Koenders *et al.*, 2014; Xiao *et al.*, 2005) and durability (Beauchemin *et al.*, 2018;
64 Levy and Helene, 2004). However, there has not been sufficient research focusing on utilizing
65 RAs in concrete for further sustainable applications, such as exploring using RAC in air
66 purification purpose by adopting the “passive strategy” within RAC (Xu *et al.*, 2018).

67 The increased usages of motor vehicles in countries such as China is causing the issue of
68 deteriorating air quality. Heavy traffic causes high concentration of hazardous pollutants such as
69 NO_x, further leading to problems in environmental deterioration and public health (Ballari *et al.*,
70 2011; Hassan *et al.*, 2010). The traditional pavement materials could not degrade air pollutants.
71 There is a practical need to develop pavement or road materials that could purify the air. Since
72 Fujishima and Honda (1972) who applied Titanium Dioxide (TiO₂) for photocatalytic
73 decomposition of water, there have been several studies (Chen and Poon, 2009; Faraldos *et al.*,
74 2016; MacPhee and Folli, 2016; Poon and Cheung, 2007; Yang, *et al.*, 2017) applying TiO₂ in

75 cementitious materials to test the photocatalytic performance. TiO₂ photocatalysis can be used in
76 products for air purification purpose (Nakata and Fujishima, 2012) by degrading air pollution
77 particles (e.g., NO_x). Various methods of applying TiO₂ in cementitious materials have also been
78 described in existing studies. For example, Guo et al. (2017) applied the nano- TiO₂ in concrete
79 surface layers using two different methods, namely intermixing and spray-coating. The mix
80 design in the study of Guo et al. (2017) was 0.75:0.25:3.0:0.3 for Portland cement, fly ash,
81 recycled coarse aggregates (RCAs), and water by mass. TiO₂ content on the concrete surface was
82 2.8% by weight, and was reduced to 2.0% after abrasion. Shen *et al.* (2015a, b) developed the
83 photocatalytic self-cleaning concrete with its surface covered by C-S-H gel and TiO₂ layer. It
84 was revealed that the surface of the self-cleaning concrete was covered by C-S-H and TiO₂ nano
85 particles around tens of nm. Mahy et al. (2019) applied TiO₂ for the photocatalytic purpose on
86 roads. It was found that the photocatalytic activity of TiO₂ reached the maximum NO degradation
87 at 53% when the TiO₂ was coated to the concrete substrates (e.g., pavement blocks) at the
88 loading rate between 10 and 12 g/m².

89 Pervious concrete has been utilized in various existing studies to address environmental issues,
90 such as flooding (Kia *et al.*, 2019), heat island effect (Liu *et al.*, 2018), and air purification by
91 applying TiO₂ as the photocatalytic agent (Asadi *et al.*, 2014). A review of these previous studies
92 applying TiO₂ in cementitious materials for air purification purpose reveals that: 1) most of these
93 existing studies applied TiO₂ in conventional cementitious products (Hunger *et al.*, 2010; Shen *et*
94 *al.*, 2015b) without utilizing RAs. However, Xu *et al.* (2018) found that RAs, due to their
95 internal porosities, could absorb more TiO₂-based catalysts compared to natural aggregates; 2)
96 limited studies have applied RAs in pervious concrete for air purification tests, although there
97 have been existing standards (China Architecture & Building Press, 2016) adopting RAs in the

98 mix design of pervious concrete; and further 3) practically, conventional concrete coated with
99 TiO_2 may have limited durability of its photocatalytic function after being exposed to rainfalls,
100 because TiO_2 coated to the surface of conventional concrete could easily be washed away by
101 rainwater There have been limited studies focusing on how the photocatalytic performance of
102 concrete pavement products could last longer, such as by introducing RAs. Extending the
103 research of Xu *et al.* (2018), one of the novelties of this study is to utilize the features of RAs due
104 to their internal voids which could absorb TiO_2 nano particles. The current study contributes to
105 developing the technical approach of enhancing both the photocatalytic performance and the
106 durability of pervious concrete containing RCAs.

107 This study aims to address the two main aforementioned issues in China (i.e., overwhelming
108 amount of C&D wastes and air pollution). The objectives of this research include: 1) comparing
109 the photocatalytic performance of RCAs and natural aggregates in different sizes when they are
110 applied with nano- TiO_2 catalysts; 2) investigating the effects of mix design parameters on
111 pervious concrete strength and further identifying the optimized mix design in order to achieve
112 the highest compressive strength in pervious concrete containing RCAs; 3) identifying the
113 optimized concentration of TiO_2 solution in terms of achieving the highest air pollutant
114 degradation rate; and 4) exploring the photocatalytic efficiency change of pervious concrete
115 containing TiO_2 -coated RA after 10-min heavy rain wash.

116 **2. Materials and methods**

117 *2.1. Concrete materials*

118 Ordinary Portland cement of C42.5 Grade was used for the mixture of pervious concrete.
119 Class II fly ash supplied by Beilun Power Plant in Ningbo China was adopted as the
120 supplementary cementitious material. Coarse aggregates sized between 5mm and 10mm were

121 adopted for both RCAs and natural coarse aggregates (NCAs) in this research. The selection of
 122 coarse aggregate size met the requirements of Technical Specification for Application of
 123 Pervious Recycled Aggregate Concrete (China Architecture & Building Press, 2016).The
 124 properties of coarse aggregates are listed in Table 1.

125 Table 1.Properties of coarse aggregate

Aggregates type	Apparent density (kg/m ³)	Bulk density (kg/m ³)	Surface dry water absorption rate (%)
RCA	1766	1115	6.4
NCA	2820	1453	0.4

126
 127 Polycarboxylic acid superplasticizer with water reduction rate at 25% was applied. TiO₂ was
 128 used as the photocatalyst in this study. The 25% water reduction rate for the superplasticizer was
 129 chosen because of the high cohesiveness of recycled aggregate concrete which generally had
 130 lower slump. It was recommended to have the water reduction rate over 20%. The
 131 superplasticizer used in this study was polycarboxylate, which was convenient for the mix design
 132 of pervious concrete containing RCAs. Table 2 displays the physical properties of TiO₂.

133 Table 2. Properties of TiO₂

Appearance	White powder
Crystal structure	Mischcrystal TiO ₂ (approximately 75% of Anatase and 25% of Rutile)
TiO₂ Content	>96
Photocatalytic efficiency	≥68

134
 135 2.2. *Preparation of TiO₂-based photocatalysts*

136 The TiO₂ concentration at 0.3% was adopted by Yang *et al.* (2017) for the photocatalytic
 137 depollution test. However, the ideal or optimized TiO₂ concentration was not identified. As a step
 138 forward from the study of Yang *et al.* (2017), the TiO₂ solution with concentration up to 0.6%
 139 was prepared by ultrasonically dispersing TiO₂ powders into distilled water. RCAs were soaked
 140 in the solution for 24 hours and then removed to be dried under the controlled temperature of

141 105°C. Fig. 1 shows the TiO₂ solution and recycled coarse aggregates (RCAs) soaked with TiO₂.

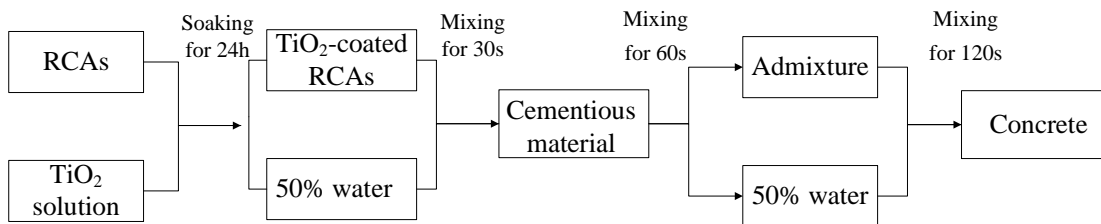


142

143 (a) TiO₂ solution with concentration at 0.6% (b) RCAs soaked with TiO₂

144 Fig. 1. Preparation of RCAs soaked with TiO₂

145 After obtaining RCAs soaked with TiO₂, the method of aggregates coated with cement pastes
146 described by Ping and Beaudoin (1992) was followed to produce pervious concrete containing
147 TiO₂-soaked RCAs. Fig. 2 describes the detailed steps of pervious concrete preparation.



148

149

150 Fig.2. Workflow of preparing pervious concrete containing TiO₂-soaked RCAs

151 According to Fig. 2 and following the mix design procedure, RCAs were initially immersed in
152 the TiO₂ solution to obtain the TiO₂-soaked RCAs for later concrete mixing. Then the natural
153 aggregates, 50% of the mixture water, and TiO₂-coated RCAs were added into the mixer. The
154 mixing time was 30s in order to have the aggregates uniformly wetted. Then the cementitious
155 materials were added to the mixer for another 60s' mixing in order to have the binder materials

156 wrapping the aggregate surface. Finally, the remaining 50% of the mixture water and the
157 superplasticizer were added to the mixer for another two minutes' mixing.

158 2.3. Orthogonal arrays and Taguchi method

159 The orthogonal array design adopting the Taguchi method (ReliaSoft, 2012) was adopted as
160 the statistical approach to identify the optimal mix design of pervious concrete according to the
161 compressive strength. Taguchi method, also known as orthogonal array design method, is a
162 broadly accepted method in design of experiment, which has proven in producing high-quality
163 outputs at subsequently low cost (Davis and John, 2018). The Taguchi Orthogonal Array is a
164 highly fractional orthogonal design based on a design matrix and allows the selection of subset
165 of combinations of multiple factors at multiple levels (ReliaSoft, 2012), for example,
166 replacement ratio of fly ash to Portland cement (FA%), the replacement ratio of RCAs to NCAs
167 (RCA%), and water-to-binder (w/b) ratio in this study. The experiments required to complete this
168 statistical test is relatively small, such as nine experimental groups in this study. The Taguchi
169 Orthogonal Array method, generally based on uniformly distributed datasets, can be combined
170 with range analysis and analysis of variance (ANOVA) to evaluate the significance of the effect
171 of each independent variable on the dependent variable. More detailed descriptions and steps to
172 conduct the Taguchi method can be found in Koschan and Antony (2006).

173 Following the Technical Specification for Application of Pervious Recycled Aggregate
174 Concrete (China Architecture & Building Press, 2016), the compressive strength of pervious
175 concrete should not be lower than 20 MPa. The compressive strength of pervious concrete should
176 be considered of fundamental importance before conducting further photocatalytic
177 environmental tests. In this study, the orthogonal array design was only applied in determining
178 the optimal mix design to achieve the required compressive strength before conducting the

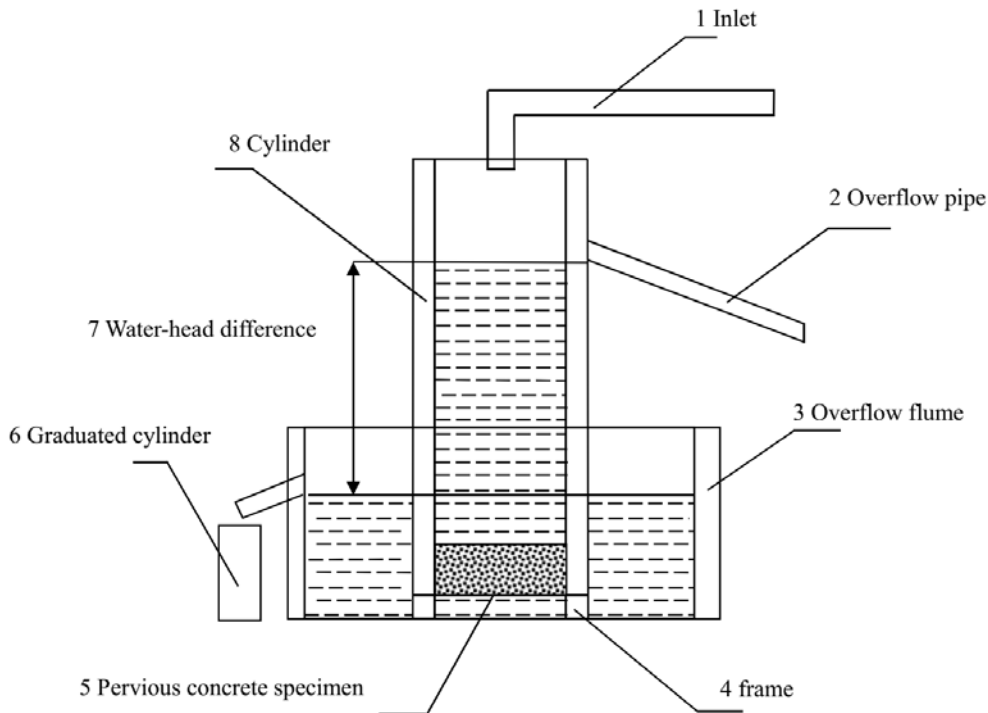
179 further photocatalytic tests.

180 The key to orthogonal design is the selection of the orthogonal table. The orthogonal table is
181 denoted as $L_n(a^p)$, where L represents an orthogonal table; n denotes the number of rows in the
182 table, which is also the number of experimental sets; p is the number of columns in the
183 orthogonal table, which is also the maximum number of factors that can be arranged in the table;
184 a indicates the number of levels taken by each factor. In this study, the orthogonal table
185 consisting of four factors and three levels denoted as $L_9(3^4)$ was defined. The four factors of w/b
186 ratio, coarse aggregate-to-binder ratio, FA%, and RCA% were selected. These three levels and
187 four factors are shown in Table 5.

188 2.4. Test facilities

189 The compressive strength of concrete specimen in this study was tested following *Standard for*
190 *Test Method of Mechanical Properties on Ordinary Concrete* (2002). The cubes with dimensions
191 of 100 mm ×100 mm×100 mm were prepared for the compressive strength test, with the loading
192 rate at 0.4 MPa/s. The permeability coefficient of pervious concrete was tested following
193 *Technical Specification for Pervious Cement Concrete Pavement* (2009). The detailed test
194 procedure is described in the following steps: (1) placing the concrete specimen in the vacuum
195 barrel, evacuating it to reach the pressure of (90 ± 1) kPa, and keeping it for 30 minutes; (2)
196 adding water to the vacuum barrel to make the water level higher than the specimen by 100 mm;
197 and (3) stopping vacuuming for 20 minutes and removing the sample from the vacuuming barrel.
198 Afterwards, following the schematic diagram illustrated in Fig. 3, the test specimen (denoted as 5
199 in Fig. 3) is placed in the cylinder (i.e., 8). The frame denoted as 4 in Fig. 3 is placed in the
200 overflow water tank (i.e., 3). Then the water inlet (i.e., 1) is opened. The water flows through the
201 cylinder (i.e., 8) and passes through the pervious concrete specimen (i.e., 5) into the overflow

202 flume (i.e., 3) until water in the cylinder (i.e., 8) starts being discharged through the overflow
 203 pipe (i.e., 2). The water inflow amount is then adjusted to balance the water inlet (i.e., 1) and the
 204 overflow pipe (i.e., 2), Finally, the water amount (denoted as Q) in the graduated cylinder (i.e., 6)
 205 and the water-head difference (denoted as h) in the steady period are recorded. The temperature
 206 (denoted as T) at the same time is also recorded.



207

208 Fig.3. Schematic diagram for the test of permeability coefficient of pervious concrete

209 The water permeability coefficient is then calculated according to Equation (1),

210
$$K_T = \frac{Q_1 \times L}{A \times h \times t} \quad \text{Equation (1)}$$

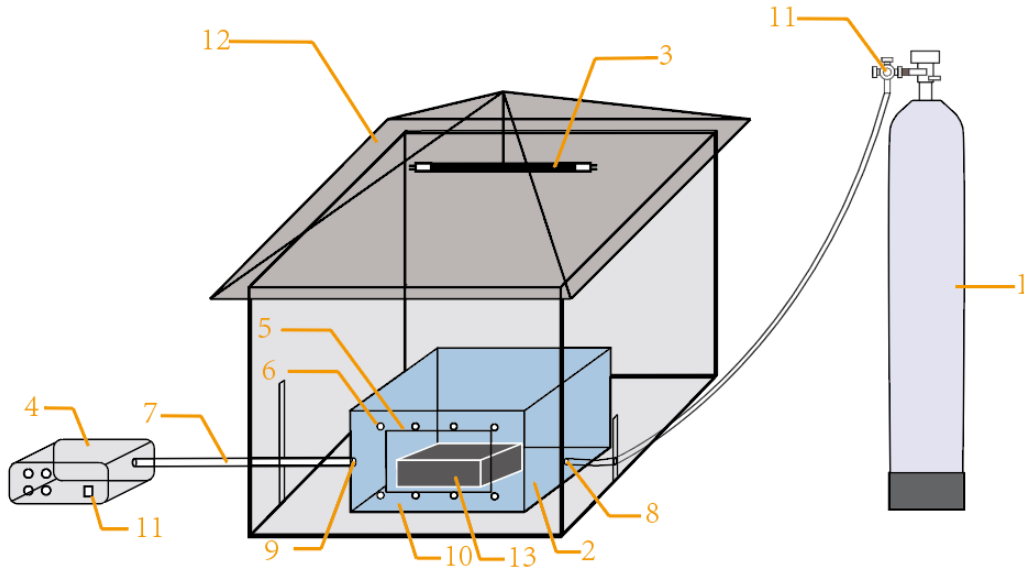
211 where K_T means the water permeability coefficient at water temperature of T ; Q_1 denotes the
 212 water amount flowing through the pervious concrete specimen by the time interval of t ; L is the
 213 thickness of the permeable concrete specimen; and A is the cross-sectional area of the specimen.

214 The microstructure of pervious concrete was observed using Phenom Pro as the desktop SEM

215 (i.e., scanning electron microscope).

216 The schematic diagram of the laboratory setup to measure NO degradation by applying the

217 TiO₂-coated pervious concrete is demonstrated in Fig.4.



218

219 1: Gas supply source; 2: Reactor; 3: UV light source; 4: Exhaust analyzer; 5: Rubber ring; 6: Screws; 7: Airway; 8:

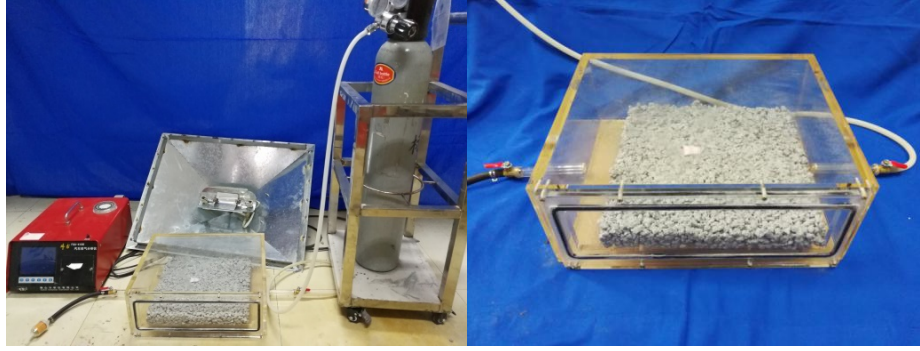
220 Air inlet; 9: Air outlet; 10: Transparent gate; 11: Valve; 12: Reflective sheltering; 13: specimen

221 Fig.4. Schematic diagram of experimental test facility to measure NO degradation

222 Following *Test Method of Photocatalytic Materials for Air Purification* (2009), the test
223 facility for the experimental measurements of NO degradation was prepared as shown in Fig.5.

224 The photocatalytic tests of degrading NO were performed following *Measurement Method for*
225 *Photolysis Performance Index of Photocatalytic Nano-Materials* (2013).

226



227

228 Fig.5. Experimental tests of applying pervious concrete specimen containing TiO₂-coated

229 RCAs to degrade NO

230 The test facility shown in Fig.5 mainly consists of NO cylinder, reactor, UV light source, the
 231 iron-made cubic box, and exhaust gas analyzer using the FGA-4100 mode.. The parameters of
 232 the exhaust gas analyzer is displayed in Table 3.

233 Table 3. Parameters of FGA-4100 mode exhaust gas analyzer

Gas for test	HC	CO	CO ₂ (%)	O ₂ (%)	NO _x (ppm)
Test method	Spectrophotometric Infrared Absorption Method		Principle of electrochemistry		
Measuring range	0-12000	0-10	0-20	0-25	0-400
Resolution ratio	1	0.01	0.1	0.1	1
Drift range /h	±4	±0.02	±0.2	±0.2	

234

235 The iron-made cubic box was manufactured as the sheltering device to prevent natural
 236 lighting coming into the reactor. The exhaust gas analyzer was used to test the NO degradation.
 237 The NO degradation rate was calculated using Equation (2):

238

$$239 \quad NO \text{ Degradation Rate} = \frac{\text{Initial concentration of NO} - \text{Final concentration}}{\text{Initial concentration}}$$

240 Equation (2)

241 The experimental procedure based on Fig. 5 and schematic diagram shown in Fig.4 is
 242 described in the following six steps: (1) placing the concrete specimen in the reactor, tightening
 243 the sealing screw, and checking the air tightness; (2) opening the outlet valve and the intake

244 valve in the reactor, opening the air pump of exhaust gas analyzer, and discharging the gas in the
245 reactor, the intake pipe, and the outlet pipe; (3) closing the air pump when the gas concentration
246 becomes constant in the exhaust gas analyzer; (4) opening the valve of the NO cylinder,
247 adjusting the size of the cylinder valve, observing the change of NO concentration on the exhaust
248 gas analyzer and waiting for the NO concentration to stay unchanged, recording the initial
249 concentration value, and then turning on the UV light source at the top of the iron-made cubic
250 box; (5) recording the NO concentration change on the exhaust gas analyzer once every three
251 minutes until the NO concentration remains unchanged within nine minutes; and (6) recording
252 the final concentration value, turning off the light source, and closing the NO cylinder value.

253 It should be noticed from Fig.4 and Fig.5 that the pervious concrete specimens under tests
254 were with the four sides unsealed. Several previous studies on the photocatalytic performance of
255 concrete specimens (e.g., Lee et al., 2014; Wang et al., 2015; and Xu et al., 2018) were also
256 found without the four sides of concrete specimens sealed to eliminate the effects from side
257 surfaces which could also be exposed to lighting. The experimental procedure was designed
258 with four sides of concrete specimens unsealed so as to allow the comparison between this study
259 and these prior studies in terms of the photocatalytic performance. As seen in Fig.4 and Fig.5, the
260 confined space with an inside UV light source could allow mainly the top surface of the concrete
261 specimen exposed to UV light, whilst the four sides of surfaces would have significantly lower
262 exposure to UV light. Recognizing the limitation regarding the four sides of specimens unsealed,
263 future work could test the side effects by also sealing the four sides of pervious concrete
264 specimens for photocatalytic experiments.

265 2.5. *Experimental study of pervious concrete exposed to rain wash*

266 It is known that the pavement's photocatalytic performance will be diminished when exposed



(a) Rain gauge

(b) Facility for rain wash

Fig.7. Rain wash facility for the experimental study of pervious concrete

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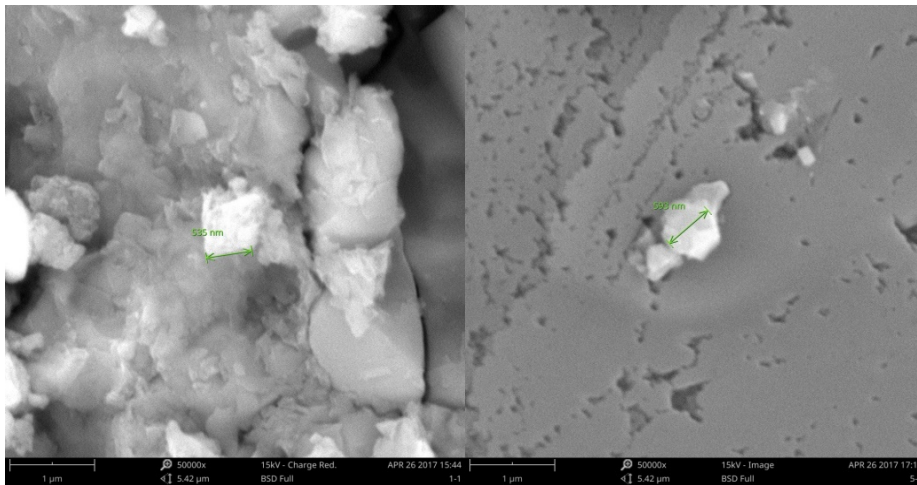
The rain gauge (Model No. J16022) is a tool for measuring the rainfall. It consists of rain inlet, a funnel, a water storage tank, and the specialized measuring cup. It was placed in the rainfall experimental test site. The rainwater fell into the rain inlet and was collected by the storage tank through the funnel. After the rainfall stopped, the water in the storage tank was measured in the measuring cup. The experimental precipitation was set at 25 cm per day or equivalent to 10.4 mm/hour, which is considered heavy storm according to United States Geological Survey (2018). The pervious concrete specimens, with surface area dimension of 300mm by 300mm and thickness of 50 mm, received the precipitation of 6 cm³ in 10 minutes during the experiment. Adopting this precipitation rate, the NO degradation rates before and after rainwater wash were obtained to measure the deterioration of photocatalytic performance of pervious concrete. The simulation of heavy storm rainfall for each test specimen was controlled at 10 min to ensure that all tests were under the same rainfall conditions. The 10-min period is considered reasonable in simulating the real-life heavy storm, which is generally with high intensity of rainfall but in a short period. Actually, the real-life heavy storm may be with a non-uniform pattern and unrealistic for comparative studies of different concrete specimens. Therefore, the current

296 experimental setup for the rainfall test is considered realistic for studying the photocatalytic
297 performance change of pervious concrete with different concentrations of TiO_2 .

298 3. Results

299 3.1. Photocatalytic performance of RCAs coated with TiO_2

300 Using the desktop SEM, the nano TiO_2 particles attached into the voids of RCAs were
301 observed as shown in Fig. 8.



302
303 Fig.8. Microstructure of observing TiO_2 particles absorbed into the internal voids of RCAs (Scale
304 at 50,000:1).

305 Fig.8 captured TiO_2 particles attached to RCAs' voids from different angles and locations
306 within pervious concrete. As can be seen in Fig.8, the porous mortar attached to the surface of
307 RCAs could absorb nano TiO_2 particles after soaking RCAs into TiO_2 solution and drying them.

308 This research started by comparing the photocatalytic performance between RCAs and NCAs.
309 As shown in Fig.9 and Fig.10, NCAs and RCAs with three different sizes (i.e., 15-20 mm, 10-15
310 mm, and 5-10 mm) were obtained to test their NO degradation performance.



(a) (b) (c)

Fig.9. RCA sized at (a)15-20 mm, (b) 10-15 mm, (c) 5-10 mm



(a) (b) (c)

Fig.10. NCA sized at (a) 15-20 mm, (b) 10-15 mm, (c) 5-10 mm

Following the procedure described in Section 2.2, these six different types of coarse aggregates were soaked in TiO_2 solution with the concentration at 0.6%.

The photocatalytic experimental facility described in Figs. 5 and 7 was applied to each type of TiO_2 -soaked aggregate to test the NO degradation rates. Table 4 lists and compares the NO degradation performance between NCAs and RCAs.

Table 4. Comparison of photocatalytic performance between RCAs and NCAs

Aggregate size	5-10 mm	10-15 mm	15-20 mm
RCA	80.6%	74.4%	71.4%
NCA	66.6%	67.7%	62.3%

It can be seen from Table 4 that both RCAs and NCAs coated with TiO_2 could degrade NO.

326 However, RCAs outperformed NCAs in its photocatalytic reaction. The superior performance of
327 RCAs was mainly due to two reasons. Firstly, the internal voids of RCAs could store TiO_2 to
328 allow more photocatalytic reactions. Secondly, TiO_2 can be more easily loaded into RCAs
329 surface because of mortar attached to its surface as shown in Fig.9. NCAs, in comparison, have a
330 less rough surface and are hence more difficult to have TiO_2 attached to them. Further comparing
331 the photocatalytic performance of the same type of aggregate with different sizes reveal more
332 findings. TiO_2 could be absorbed either into the internal voids or surface of RCAs. RCAs with
333 smaller sizes would have more internal porosities and higher specific surface area to absorb more
334 TiO_2 . Therefore, as the size of RCAs increases, the photocatalytic performance would decrease
335 due to the reduced internal porosities and lower specific surface area. Different from RCAs,
336 NCAs have little internal porosity. The TiO_2 particles could only be attached to the surface of
337 NCAs, hence the effects of NA size on the photocatalytic performance turned out less significant.
338 As seen in Table 4, the photocatalytic performance for NCAs sized between 5-10mm and 10-
339 15mm was least significant, although there was a minor reduction of photocatalytic performance
340 when the NCA size reached 15-20 mm.

341 3.2. *Mix design of pervious concrete adopting RCAs*

342 According to *Technical Specification for Application of Pervious Recycled Aggregate*
343 *Concrete* (China Architecture & Building Press, 2016), the replacement ratio of RCAs to NCAs
344 (RCA%) should not be lower than 30%, water-to-binder (w/b) ratio should be between 0.25 and
345 0.35, and the replacement ratio of fly ash to Portland cement (FA%) should not be higher than
346 30%. The w/b ratio and coarse aggregate-to-binder ratio (a/b) are considered critical factors
347 affecting the mechanical properties of pervious concrete (Shi et al., 2016). Therefore, the four
348 factors (i.e., RCA%, w/b , FA%, and a/b) were decided. Following the orthogonal table in the

349 form of $L_9 (3^4)$, three levels were confirmed for the four factors. Further based on the
 350 researchers' own prior trials, the levels for each factor shown in Table 5 were determined by the
 351 research team. For example, should not be lower than 0.27 to maintain the workability of
 352 pervious concrete containing RCAs.

353 Table 5. Parameters defined in the orthogonal experimental design

Level	Independent variables			
	(A) <i>w/b</i>	(B) <i>a/b</i>	(C) FA%	(D) RCA%
(a)	0.27	3	5%	30%
(b)	0.3	3.3	10%	50%
(c)	0.35	4	20%	70%

354
 355 Based on the orthogonal design defined in Table 5, nine experimental tests were performed
 356 covering different combinations of these four independent variables. The compressive strength
 357 and photocatalytic performance measured by NO degradation rates were tested and summarized
 358 in Table 6.

359 Table 6. Experimental arrangement and range analysis for comprehensive strength of pervious
 360 concrete

Experiment No.	(A)	(B)	(C)	(D)	Index	
	<i>w/b</i>	<i>a/b</i>	FA%	RCA%	Compressive strength (MPa)	NO Degradation rate
1#	0.27(a)	3(a)	5%(a)	30%(a)	9.0	47%
2#	0.27(a)	3.3(b)	10%(b)	50%(b)	6.8	62%
3#	0.27(a)	4(c)	20%(c)	70%(c)	2.2	38%
4#	0.3(b)	3(a)	10%(b)	70%(c)	7.1	14%
5#	0.3(b)	3.3(b)	20%(c)	30%(a)	4.4	18%
6#	0.3(b)	4(c)	5%(a)	50%(b)	7.4	52%
7#	0.35(c)	3(a)	20%(c)	50%(b)	19.8	33%
8#	0.35(c)	3.3(b)	5%(a)	70%(c)	13.8	10%
9#	0.35(c)	4(c)	10%(b)	30%(a)	9.2	17%
Range Analysis	Index	Index	Index	Index	---	---
K_I	6.000	11.967	10.067	7.533		
K_{II}	6.300	8.333	7.700	11.333		

K_{III}	14.267	6.267	8.800	7.700		
Y	8.267	5.700	2.367	3.800		

361

362 The range analysis of orthogonal test was performed following the procedure described in Xu
 363 (2015). In Table 6, K_i denotes the average comprehensive strength for a certain variable at Level
 364 i (e.g., I , II , or III). By comparing and evaluating the index values (i.e., compressive strength) at
 365 K_i , the optimal level of variables can be confirmed. The parameter Y , as shown in Table 6, is
 366 computed following Equation (3),

367
$$Y \text{ (MPa)} = \max\{K_I, K_{II}, K_{III}\} - \min\{K_I, K_{II}, K_{III}\} \quad \text{Equation (3)}$$

368 The parameter Y shows the effect of variables on the compressive strength. A high Y value of
 369 corresponding to a certain variable (e.g, w/b) means that this variable has a relatively strong
 370 effect on the compressive strength. The significance of each independent variable on the
 371 compressive strength was tested using ANOVA (i.e., analysis of variance) as shown in Table 7.
 372 The three different key threshold values (i.e., K_I , K_{II} , and K_{III}) linked to the different variables are
 373 displayed in Figs.11-13.

374 Following the experimental data collected in Table 6, a F -test based on ANOVA was
 375 performed to evaluate the impact of each independent variable on the compressive strength of
 376 pervious concrete. As displayed in Table 7, the related F value is a key parameter of ANOVA.
 377 The F value was computed following Equation (4),

378
$$F = \frac{S_A / f_A}{S_e / f_e} \quad \text{Equation (4)}$$

379 where S_A denotes the sum of squared deviations of factors; f_A is the degree of freedom of
 380 factors; S_e is the sum of squared deviations of experimental errors; and f_e means the degree of
 381 freedom of experimental errors. Using the F -test to evaluate the effects of independent mix
 382 design variables on concrete strength can be found in several existing studies (e.g., Jin *et al.*,

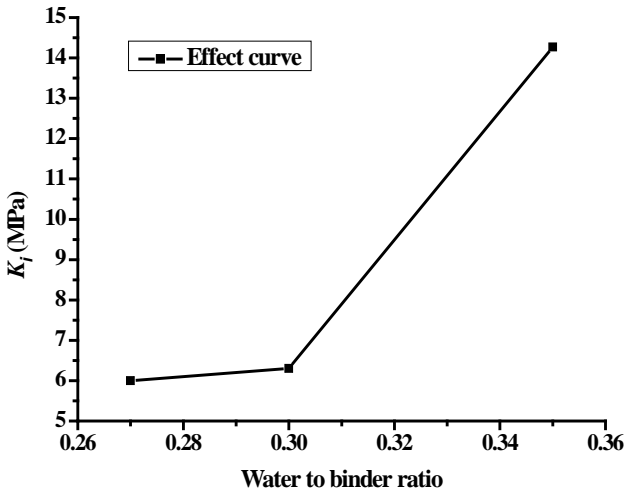
383 2018b).

384 The null hypothesis was that the given independent variable did not have a significant effect
385 on pervious concrete strength. The null hypothesis would be declined if the obtained F value is
386 equal to or higher than the critical F value at defined levels of significance (i.e., 0.05, 0.1, and
387 0.25 in Table 7). A higher F value would indicate that the given independent variable had a more
388 significant impact on the compressive strength of pervious concrete.

389 Table 7. ANOVA for comprehensive strength

Indepen- dent variable	Sum of squares	Degrees of freedom	Variance	F value	p value	F value at defined level of significance	Evaluation of significance
w/b	131.6792	2	65.84	15.74	0.01		Most significant
a/b	49.9832	2	24.99	5.97	0.06	$F_{0.05}(2,4)=6.94$	Significant
RCA%	27.7152	2	13.86	3.31	0.14	$F_{0.1}(2,4)=4.32$	Less significant
FA%	8.3672	2	4.184	1.00		$F_{0.25}(2,4)=2.00$	Least significant
Error	8.3672	2	4.18				
Correc- tion	16.7344	4	4.18				
Error Total	226.112						

390
391 As seen in Table 7, there are three threshold F values corresponding to the three different
392 levels of significance. The three different threshold values define the significance of each
393 independent variable in one of the four different categories (i.e., “most significant” if the F value
394 is higher than 6.94; “significant” if the computed F value is between 4.32 and 6.94; “less
395 significant” if the F value is between 2.00 and 4.32; and “least significant” if the F value is lower
396 than 2.00. It can be seen from Table 7 that w/b (i.e., water-to-binder) ratio has the highest impact
397 on concrete strength, followed by a/b (i.e., aggregate-to-binder ratio), replacement ratio of RCAs,
398 and fly ash percentage. As seen in Fig. 11, the compressive strength of pervious concrete on Day
399 28 increased with w/b ratio.



400

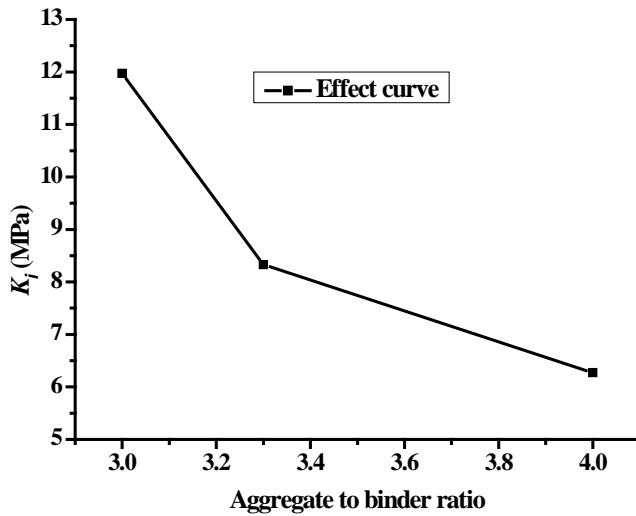
401 Note: the definition of K_i in Figs.11-13 is consistent with what has been defined in Table 6.

402 Fig. 11. 28-day compressive strength of pervious concrete at different w/b ratios

403 The trend of compressive strength change with w/b ratio in pervious concrete turned out
 404 different compared to that in conventional concrete due to the fact that the strength growth in
 405 pervious concrete does not follow Bolomey's equation (Bolomey, 1927). Instead, pervious
 406 concrete is designed to have voids to allow moisture to permeate through it. The higher porosity
 407 within pervious concrete causes strength reduction. The strength developed within pervious
 408 concrete is based on contact points connected within its internal structure and the bonding
 409 strength at connections (Zhong and Wille, 2016). Pervious concrete produced in this research
 410 was based on the method of coating aggregates with cement paste (Ping and Beaudoin, 1992). A
 411 lower w/b ratio would cause insufficient hydration of cementitious materials, leading to lower
 412 bonding strength at connections. RCAs would absorb more moisture than NCAs in this study,
 413 causing inadequate hydration of cementitious materials at contact point connections and resulting
 414 in lower strength developed.

415 The effect of a/b ratio on concrete compressive strength is shown in Fig. 12. A negative
 416 relationship between a/b ratio and compressive strength can be observed. As indicated in Fig.12,

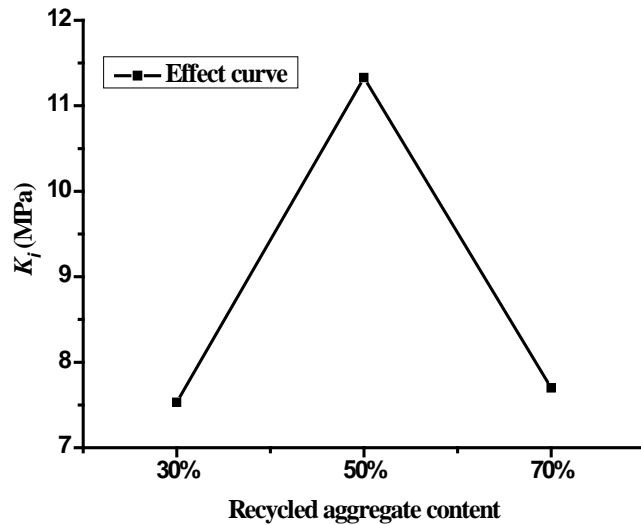
417 when a/b ratio is higher than 4.0, there would be insufficient cementitious materials covering
418 aggregate surfaces, causing their inadequate hydration which further leads to lower bonding
419 strength at contact points.



420

421 Fig.12. 28-day compressive strength of pervious concrete at different a/b ratios

422 The effect of RCA% (i.e., replacement ratio of RCAs to NCAs) on concrete strength is
423 illustrated in Fig. 13. It is seen in Fig. 13 that the compressive strength of pervious concrete
424 would first increase with RA% until it reaches 50%, and then decrease as RA% continues
425 increasing.



426

427 Fig. 13. 28-day compressive strength of pervious concrete at different replacement ratios of
 428 RCAs

429 Consistent to the earlier study performed by Jin *et al.* (2018a), Xu and Sun (2011), and Xu *et*
 430 *al.* (2018), lower replacement ratios of RCAs could increase the concrete strength due to the fact
 431 that RAs had their positive effects (e.g., internal curing) in developing concrete strength. This
 432 positive effect of RAs on concrete strength was not affected by adding TiO_2 particles (Xu and
 433 Sun, 2011). However, as the RA replacement ratio increases, the negative effects of RAs would
 434 outweigh their positive impacts, due to RA's lower quality in terms of their physical properties
 435 and weak interfacial transition zones identified by earlier studies (Lei *et al.*, 2018; Li *et al.*, 2017;
 436 Limbachiya *et al.*, 2000). As a result, a higher percentage (i.e., over 50%) of RCAs replacing
 437 NCAs would decrease concrete strength.

438 The optimized mix design for pervious concrete containing TiO_2 -coated RCAs is therefore
 439 identified in order to achieve the highest compressive strength. Table 8 lists the optimized mix
 440 design.

441 Table 8. Optimized mix design of pervious concrete in this study

<i>w/b</i>	<i>a/b</i>	FA%	RCA%	Compressive strength on Day 28 (MPa)	Water permeation coefficient (mm/s)	NO degradation rate (%)
0.35	3	5%	50%	21.6	12.5	37.4

442

443 3.3. *Effects of TiO₂ concentration on NO degradation rates of photocatalytic pervious*
444 *concrete*

445 The optimized mix design parameters identified in Table 8 were then adopted to produce
446 pervious concrete with RCAs containing nano particles from different concentrations of TiO₂
447 solution. The photocatalytic performance in terms of NO degradation rates were tested and
448 measured both before and after the 10-min heavy rainwater wash adopting the precipitation rate
449 described in Section 2.5. Six different concentrations (i.e., 0%, 0.1%, 0.2%,0.3%, 0.4%, and
450 0.5%) of TiO₂ solution were studied in order to identify the optimized concentration for
451 achieving the highest photocatalytic performance. Data collected from photocatalytic tests are
452 summarized in Table 9.

453 Table 9. Photocatalytic performance of pervious concrete containing RCAs soaked at different
454 concentrations of TiO₂ solution

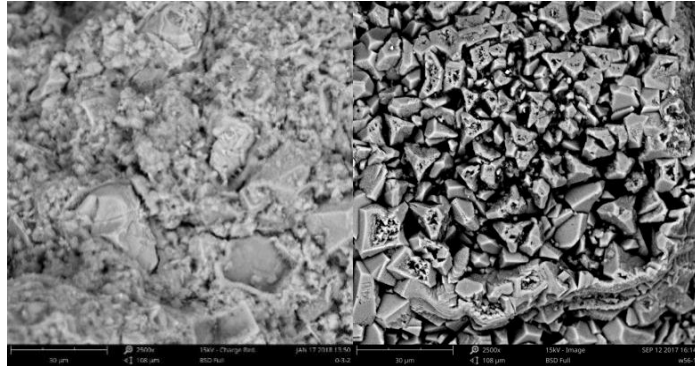
Test No.	TiO ₂ solution concentration	NO degradation rate before rainwater wash	NO degradation rate after 10-min rainwater wash
T-0	0	0	0
T-1	0.1%	61.4%	40%
T-2	0.2%	63.6%	44.30%
T-3	0.3%	70%	49.60%
T-4	0.4%	52.2%	41.50%
T-5	0.5%	42.2%	31.10%

455

456 The highest NO degradation rate was achieved at 70%. Compared to the photocatalytic
457 performance of concrete specimens containing TiO₂ nano particles in some previous studies, e.g.,

458 NO degradation rate at 17.6% in Xu et al. (2018), 32%-56% in Mahy et al. (2019), and 62% in
459 Yang et al. (2019), this study achieved the highest NO degradation rate by adopting TiO₂-soaked
460 RCAs in the mix design of pervious concrete.

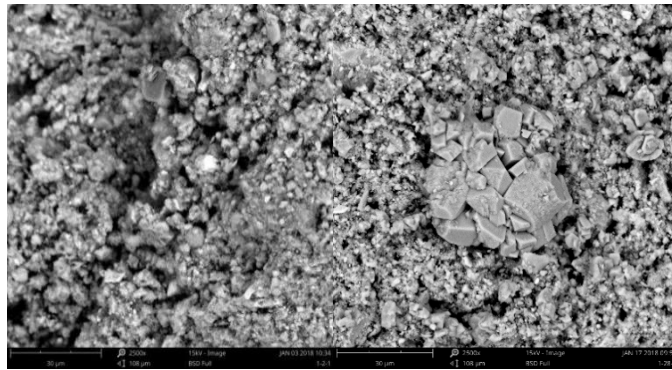
461 It is also indicated from Table 9 that the photocatalytic performance of pervious concrete
462 containing TiO₂-coated RCAs did not always increase with the concentration of TiO₂ solution.
463 Instead, there was an optimized TiO₂ solution concentration identified at 0.3%. The highest NO
464 degradation rate was achieved at 70% when 0.3% concentration of TiO₂ solution was used to
465 soak RCAs. Compared to the photocatalytic performance of ready-mix concrete in the study of
466 Xu *et al.* (2018) where the NO_x degradation rate was below 20%, applying TiO₂-coated RCAs
467 could significantly increase the air pollutant degradation rate. The photocatalytic performance of
468 pervious concrete would be decreased when the concentration of TiO₂ solution was higher than
469 0.3%. This could be due to the fact that the TiO₂ concentration over 0.3% decreased the
470 permeability of pervious concrete as evidenced by the experimental tests. In this study, following
471 the experimental test procedure illustrated in Fig.3 and Equation (1), the permeability
472 coefficients for pervious concrete specimens with TiO₂ concentrations at 0.3%, 0.4%, and 0.5%
473 were 15.45 mm/s, 14.78 mm/s, and 13.36 mm/s respectively. It was indicated that the increased
474 TiO₂ concentration reduced the permeability coefficients of pervious concrete specimens, further
475 decreasing the exposure of the photocatalytic nano-materials to UV light. As a result, the
476 photocatalytic performance was lowered. To further validate the effects of TiO₂ concentration on
477 pervious concrete's photocatalytic performance, the micro-structures of pervious concrete with
478 RCAs containing TiO₂ particles from the five different concentrations of TiO₂ solution are
479 captured in Fig. 14.



480

481

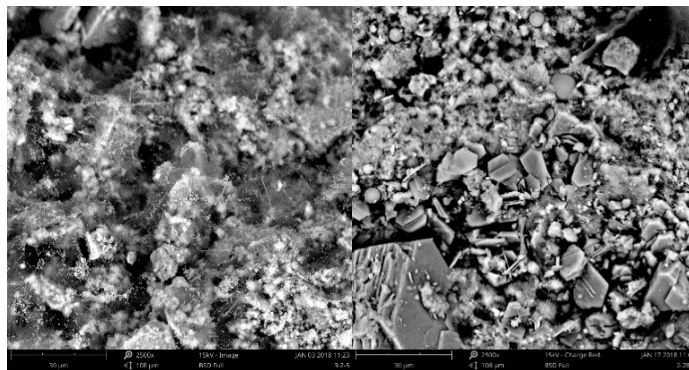
(a) Concrete microstructure observed with RCAs soaked in 0% concentration of TiO₂ solution



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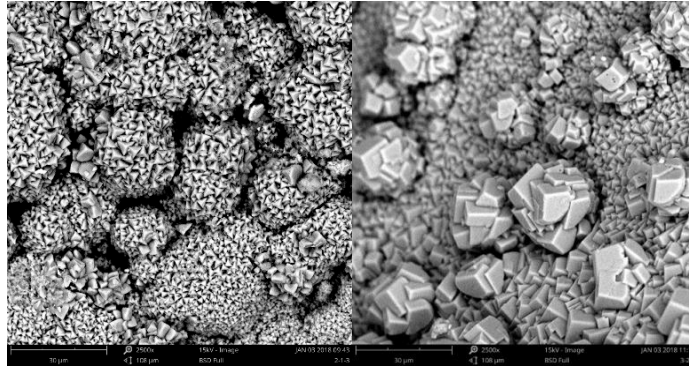
(b) Concrete microstructure observed with RCAs soaked in 0.1% concentration of TiO₂ solution



484

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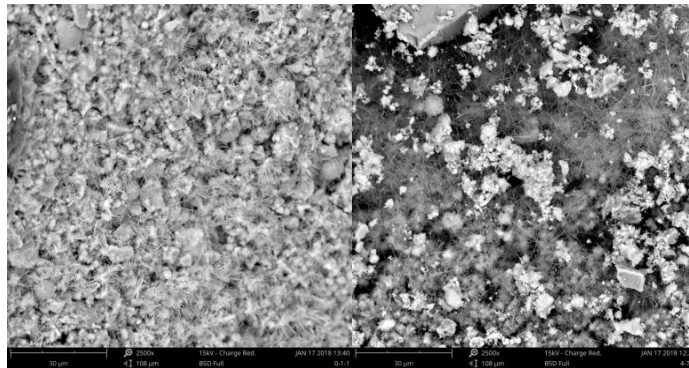
(c) Concrete microstructure observed with RCAs soaked in 0.2% concentration of TiO₂ solution



486

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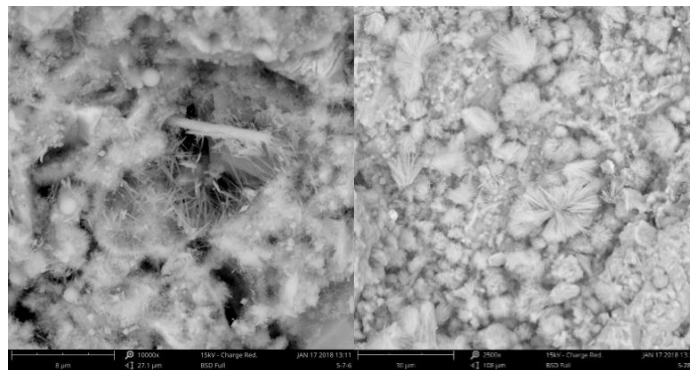
(d) Concrete microstructure observed with RCAs soaked in 0.3% concentration of TiO₂ solution



488

489

(e) Concrete microstructure observed with RCAs soaked in 0.4% concentration of TiO₂ solution



490

491

(f) Concrete microstructure observed with RCAs soaked in 0.5% concentration of TiO₂ solution

492

493

Fig. 14. Micro-structure of pervious concrete containing TiO₂-coated RCAs soaked in different concentrations of TiO₂ solution

494

495

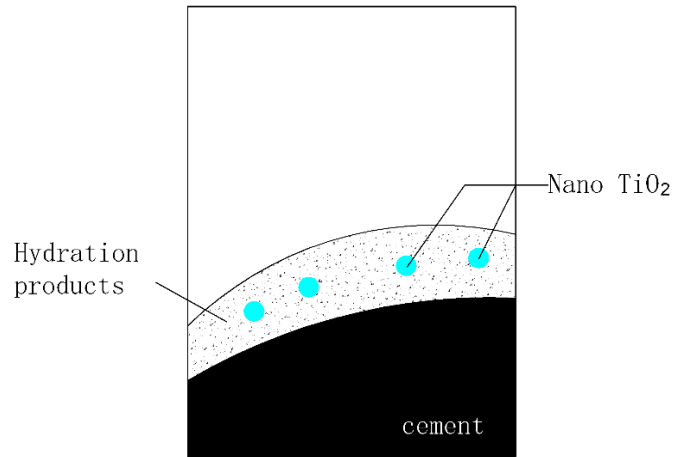
496

It can be observed from Fig. 14 that when using RCAs after being soaked into 0% concentration of TiO₂ solution, the cement hydration products appeared large but not solid. According to Fig. 14-b) and c), the nano feature of TiO₂ particles formed cores during cement

497 hydration, increasing the forming rates of cement crystals and also reducing the size of hydration
498 products(Chandrappa and Biligiri, 2016). When the 0.3% concentration of TiO_2 solution was
499 used to soak RCAs, TiO_2 not only accelerated the formation of dense hydration layer on cement
500 surfaces, but also formed globular-shaped hydration product (Chen *et al.*, 2011) outside the
501 hydration layer as seen in Fig.14-c). However, as the concentration of TiO_2 solution continued
502 increasing from 0.3%, the calcium meteorite (i.e., AFt) increased rapidly and consumed TiO_2
503 particles, and further reducing the photocatalytic effect of concrete.

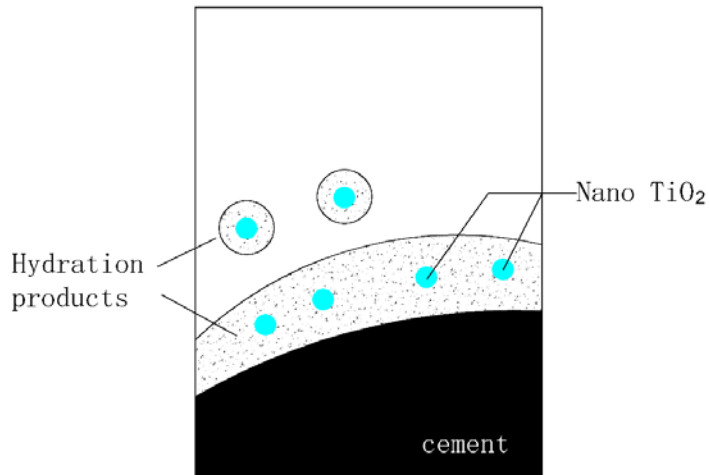
504 The effect of the concentration of TiO_2 solution on cement hydration displayed in Fig. 14 can
505 be further illustrated in Fig. 15. When the concentration of TiO_2 solution was lower than the
506 optimized value (i.e., 0.3%), the hydration layer formed outside the cement surface. The globular
507 hydration products would be formed when the concentration was close to 0.3%. Further
508 increasing the concentration of TiO_2 solution would form more TiO_2 -driven cores, and even
509 create a secondary layer by connecting these cores. The calcium meteorite could also be formed
510 as a result. It can be further seen in Fig. 14-f) and Fig.15-c) that some TiO_2 -driven whiskers were
511 formed after soaking RCAs in higher concentration of TiO_2 solution. These whiskers could be
512 found in both the surface and inner of RCAs. RCAs could provide the internal voids to
513 accommodate whiskers and further enhancing concrete strength growth (Xu *et al.*, 2018).

514



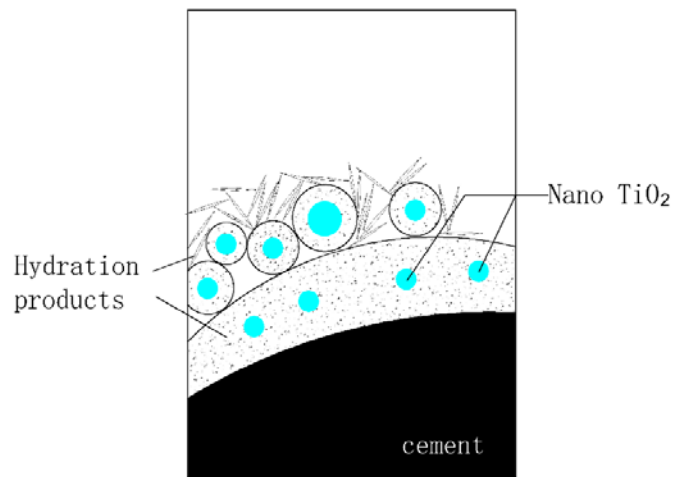
515
516

(a) Concentration of TiO₂ solution lower than 0.3%



517
518

(b) Concentration of TiO₂ solution at the optimized value of 0.3%



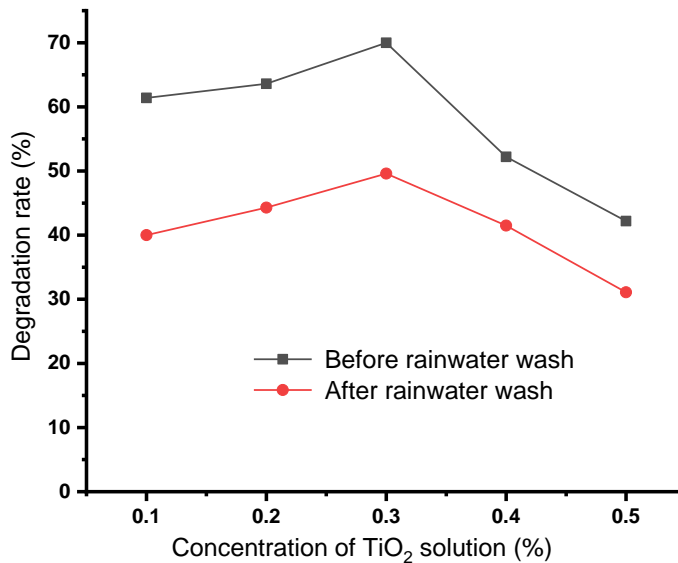
519
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(c) Concentration of TiO₂ solution higher than 0.3%

521 Fig. 15. Illustration of cement hydration product formation at different concentrations of TiO₂

522 solution

523 The photocatalytic performance of pervious before and after the 10-minute heavy rainwater
524 wash was compared as seen in Fig. 16.



525

526 Fig. 16. NO degradation rates of pervious concrete specimens before and after 10-min heavy
527 rainwater wash

528 According to Fig. 16, the photocatalytic performance of all specimens was reduced after 10-
529 min rainwater wash, with the reduction of degradation rates of NO ranging from 20% to 35%.
530 Rainwater wash reduced the nano TiO₂ particles. It was also found that pervious concrete with
531 RCAs soaked in 0.3% TiO₂ solution still achieved the highest degradation rate (i.e., 49.6%) after
532 10-min rainwater wash.

533 4. Conclusion

534 This research focused on applying the TiO₂-soaked RCAs in pervious concrete by
535 investigating its impacts on concrete mechanical strength as well as photocatalytic performance.
536 The effects of nano TiO₂ particles on cement hydration were investigated through observation of

537 concrete microstructure. Main findings are summarized below:

- 538 • Compared to NCAs, the porosity and rough surface of RCAs enabled nano TiO_2 particles
539 being attached to them and further improved the photocatalytic effect. Smaller-sized RCAs,
540 due to their higher specific surface area, would have better photocatalytic performance.
- 541 • The optimized mix design of pervious concrete adopting TiO_2 -soaked RCAs was identified
542 through orthogonal experimental design to achieve the highest compressive strength: water-
543 to-binder ratio at 0.35, aggregate-to-binder ratio at 3, fly ash percentage at 5%, and the
544 replacement ratio of RCAs to NCAs at 50%.
- 545 • RCAs were found with its positive effects on improving pervious concrete strength when its
546 replacement ratio to NCAs were below 50%.
- 547 • Six different concentrations of TiO_2 solution were compared of their effects on the
548 photocatalytic effects of pervious concrete. The optimized concentration was identified at
549 0.3% in order to achieve the highest NO degradation rate at 70%.
- 550 • Microstructure observation revealed that TiO_2 particles formed cores and enhanced the
551 formation of globular cement hydration products when pervious concrete was produced from
552 RCAs soaked in 0.3% TiO_2 solution. But when the concentration was higher than 0.3%,
553 especially when reaching 0.5%, more TiO_2 -driven cores and even whiskers would be formed,
554 creating a secondary hydration layer and more calcium meteorites, the latter of which turned
555 out reducing the concrete photocatalytic performance.
- 556 • After a 10-minute heavy rainwater wash, the pervious concrete specimens with RCAs soaked
557 in 0.3% concentration of TiO_2 solution still displayed the highest NO degradation rate at
558 nearly 50%, indicating a more durable photocatalytic performance.

559 This research addressed the gaps in applying concrete wastes for environmental protection by

560 initiating a prototype of photocatalytic pervious concrete. RCAs were found with superior
561 performance in absorbing photocatalysts (i.e., TiO₂) for air purification purpose. The study also
562 addressed the issue of durability of photocatalyst concrete under rain wash from the practical
563 perspective, indicating that applying RCAs had multiple potential advantages including reusing
564 wastes as well as improving photocatalytic performance with better durability. The
565 photocatalytic pervious concrete products may require higher initial cost. Preparing RCAs coated
566 with photocatalyst nano particles may also ask extra effort in the concrete mixture process. The
567 economical, environmental, and technical aspects of the photocatalytic pervious recycled
568 aggregate concrete need to be considered in a holistic approach.

569 Future research would also extend the current research in applying photocatalysts-coated
570 RCAs in self-cleaning concrete for air purification uses. More applications of concrete members
571 can be developed with photocatalytic functions, such as precast concrete components. It should
572 be noticed that the application of pervious concrete containing photocatalysts-coated RCAs is not
573 limited to air purification, but also water purification. More future studies could investigate the
574 water purification performance of pervious concrete by utilizing RCAs coated with
575 photocatalytic nano materials.

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