

広島大学学術情報リポジトリ
Hiroshima University Institutional Repository

Title	Effects of porous ceramic roof tile waste aggregate on strength development and carbonation resistance of steam-cured fly ash concrete
Author(s)	Ogawa, Yuko; Phuong Trinh Bui, ; Kawai, Kenji; Sato, Ryoichi
Citation	Construction and Building Materials , 236 : 117462
Issue Date	2020-03-10
DOI	10.1016/j.conbuildmat.2019.117462
Self DOI	
URL	https://ir.lib.hiroshima-u.ac.jp/00050512
Right	© 2020. This manuscript version is made available under the CC-BY-NC-ND 4.0 license http://creativecommons.org/licenses/by-nc-nd/4.0/ This is not the published version. Please cite only the published version. この論文は出版社版ではありません。引用の際には出版社版をご確認、ご利用ください。
Relation	



1 Title:
2 Effects of porous ceramic roof tile waste aggregate on strength development and carbonation
3 resistance of steam-cured fly ash concrete
4
5 Authors:
6 Yuko Ogawa (corresponding author)
7 ogaway@hiroshima-u.ac.jp
8 Department of Civil and Environmental Engineering, Hiroshima University
9 1-4-1, Kagamiyama, Higashi-Hiroshima city, Hiroshima 739-8527, Japan
10
11 Phuong Trinh Bui
12 buiphuongtrinh@hcmut.edu.vn
13 Department of Construction Materials, Ho Chi Minh City University of Technology, VNU-HCM
14 268 Ly Thuong Kiet, Ho Chi Minh city, Vietnam
15
16 Kenji Kawai
17 kkawai@hiroshima-u.ac.jp
18 Department of Civil and Environmental Engineering, Hiroshima University
19 1-4-1, Kagamiyama, Higashi-Hiroshima city, Hiroshima 739-8527, Japan
20
21 Ryoichi Sato
22 sator@hiroshima-u.ac.jp
23 Hiroshima University
24 1-4-1, Kagamiyama, Higashi-Hiroshima city, Hiroshima 739-8527, Japan

25 Abstract: (95/100 words)

26 This study aims to investigate the effects of porous ceramic waste coarse aggregate (PCWA), which
27 was used as an internal curing agent, on the compressive strength, shrinkage, and carbonation
28 resistance of steam-cured concrete using fly ash (FA) at the replacement ratios of 0%, 20%, and
29 40% by mass. The PCWA replacement ratios amounted to 0%, 10%, and 20% of the coarse
30 aggregate volume. The results showed that the PCWA significantly improved compressive strength
31 and carbonation resistance of concrete using 40% FA, whereas it could increase the drying
32 shrinkage of the steam-cured concrete using 40% FA.

33

34 Keywords: Roof tile waste, internal curing, fly ash, steam-cured concrete, carbonation resistance

35

36 1 Introduction

37 Fly ash (FA) is a by-product in coal fired power plants, and its utilization is essential to decrease the
38 environmental impact of the cement concrete industry and conserve resources. Therefore, FA has
39 been widely used in construction as a mineral admixture for concrete, a mineral filler for asphalt
40 concrete, a sub-base for pavement layers, and for similar applications. In the construction field, FA
41 is well-known as a useful supplementary cementitious material [1]. The application of FA as a
42 substitute for Portland cement can not only reduce the CO₂ emissions from cement production but
43 also enhance the properties of concrete, such as workability, long-term strength, and chemical
44 resistance. However, concrete including low-calcium FA as a substitute for Portland cement has
45 lower early-age strength and slower strength development than that without FA owing to the slow
46 FA reaction. This lower early-age strength can prevent the wider application of FA as a
47 cementitious material. To increase its applications, the utilization of FA in pretensioned prestressed
48 concrete has been investigated [2–5]. The manufacture process of pretensioned prestressed concrete

49 generally involves steam curing to obtain the required early-age strength for prestressing, and the
50 steam-cured concrete members are demolded and exposed to the air at an earlier age than those
51 cured at the normal temperature. Lothenbach et al. reported that concrete cured at the higher
52 temperature has lower long-term strength because of its higher porosity due to the denser inner
53 C-S-H and the decrease in the ettringite content at higher temperature [6]. It was also reported that,
54 compared with the standard curing (i.e., curing at 20 or 27 °C), steam curing causes concrete to
55 become more porous, resulting in lower durability and higher sorptivity [4, 7, 8]. Some researchers
56 showed that the supply of water/moisture to the demolded concrete after steam curing could
57 effectively improve concrete strength [9]. Meanwhile, Zou et al. reported that the performance of
58 steam-cured concrete, including permeability and compressive strength, decreased due to steam
59 curing followed by water curing [10]. It is debatable whether supplying water to concrete after
60 steam curing is effective at enhancing the qualities of concrete.

61 Internal curing was devised as a method for reducing the autogenous shrinkage in high-performance
62 concrete. According to Lura [11], the internal curing effect obtained using saturated porous
63 aggregate was first presented in 1967 by Aroni and Polivka, and the method has been developed as
64 a new technique that supplies water from the internal curing agent after mixing. Later, the ACI
65 Committee 308 defined “internal curing” as the process by which the hydration of cement occurs
66 because of the availability of additional internal water that is not part of the mixing water [12].
67 Lightweight aggregates and superabsorbent polymers are well-known as internal curing agents that
68 reduce early-age shrinkage. Philleo was the first to propose the use of lightweight fine aggregate as
69 an internal curing agent [13]. The internal curing effect of lightweight aggregate has been
70 investigated [14–17], and Nie et al. indicated recently that internal curing with lightweight
71 aggregate improved the performance of heat-cured concrete [9]. Jensen and Hansen were the first to
72 propose a superabsorbent polymer as an internal curing agent [18], and later, many researchers

73 investigated the use of such polymers as internal curing agents in concrete [19–21]. Besides these
74 two materials, one natural and the other artificial, some by-products or recycled materials such as
75 bottom ash and recycled aggregate have been investigated as internal curing agents [22–24].
76 Moreover, a waste aggregate derived from roof tiles was reported as an effective internal curing
77 material [25]. In the north part of Chugoku district in Japan, it is estimated that approximately
78 10,000 tons of roof tiles, called *Sekisyu Kawara*, were discarded as waste in 2015 alone owing to
79 thermal cracking during the cooling process [26]. To obtain a roof tile with the high freezing and
80 thawing resistance required in the cold weather region, *Sekisyu Kawara* is produced by sintering
81 clay at high temperatures exceeding 1200 °C, higher than the temperature applied in the production
82 of other roof tiles that are produced for use in warm regions in Japan. Suzuki et al. studied the
83 effects of *Sekisyu Kawara*, which has relatively high water absorption and relatively low crushing
84 values, as an internal curing agent, and found that the roof tile waste can develop the compressive
85 strength and reduce the autogenous shrinkage of high-performance concrete with a very low
86 water-to-binder ratio of 0.15 [25, 27]. Sato et al. used roof tile waste as the internal curing agent in
87 slag concrete, and reported that this slag concrete had higher compressive strength and denser
88 structure than that without the roof tile waste as the internal curing agent [28]. It has also been
89 reported that internal curing using roof tile waste can effectively increase the compressive strength
90 of high-volume FA concrete cured at 20 °C [29]. Moreover, the properties of steam-cured FA
91 concrete including porous ceramic roof tile waste were investigated in some earlier works [30–32].
92 It was reported that addition of roof tile waste at a replacement ratio of 10% increased the
93 compressive strength and carbonation resistance of steam-cured FA concrete [30, 32], whereas that
94 of 20% significantly decreased the autogenous shrinkage in steam-cured FA concrete [31].
95 However, these results have not been discussed sufficiently yet.
96 The purpose of this study is to investigate the effect of roof tile waste as an internal curing agent on

97 the quality of steam-cured FA concrete by providing additional data and furthering the discussions
98 of the previous literature [30-32]. In this study, pre-saturated roof tile waste replaced 10% or 20%
99 of the coarse aggregate volume in steam-cured high strength FA concrete, and its effects on the
100 compressive strength, shrinkage, and carbonation resistance of the concrete were investigated.

101

102 2 Experimental procedures

103 2.1 Materials

104 High-early-strength Portland cement meeting the standard values of the Japanese Industrial
105 Standards JIS R 5210 (Portland cement) [33] was used in this study. Low-calcium FA meeting the
106 standard values of Type II as per JIS A 6201 (Fly ash for use in concrete) [34] was also used as
107 cementitious material. The chemical composition and physical properties of these cementitious
108 materials are listed in Table 1.

109

110 Table 1 Chemical composition and physical properties of cementitious materials used in this work

	High-early-strength Portland cement	Low-calcium FA
Notation	C	F
SiO ₂ (%)	20.15	59.20
Al ₂ O ₃ (%)	4.80	23.87
Fe ₂ O ₃ (%)	2.71	5.43
CaO (%)	65.32	2.38
MgO (%)	1.15	1.01
SO ₃ (%)	3.08	0.27
Na ₂ O (%)	0.16	0.30

K ₂ O (%)	0.40	0.87
Loss on ignition (%)	1.37	2.7
Density (g/cm ³)	3.14	2.33
Blaine fineness (cm ² /g)	4490	3200

111

112 Crushed quartz-porphry was used as the conventional fine and coarse aggregates in the concrete.
 113 These fine and coarse aggregates met the standard values for manufactured sand and crushed stone
 114 as per JIS A 5005 (Crushed stone and manufactured sand for concrete) [35]. Porous ceramic waste
 115 aggregate (PCWA) derived from crushed roof tile waste was also used as an internal curing agent,
 116 partially replacing the coarse aggregate. PCWA was used in a saturated surface-dry condition after
 117 7 d of water immersion. Table 2 lists the physical properties of these aggregates. The crushing
 118 values of the coarse aggregates based on BS 812-110 (Testing aggregates. Methods for
 119 determination of aggregate crushing value) [36] are also listed in Table 2. The crushing value of the
 120 PCWA is approximately twice as high as that of the conventional coarse aggregate.

121

122 Table 2 Physical properties of aggregates

	Crushed quartz-porphry		Porous ceramic aggregate
	for fine aggregate	for coarse aggregate	for coarse aggregate
Notation	S	G	PCWA
Density (g/cm ³)	2.60	2.62	2.26
Water absorption (%)	1.16	0.62	8.70
Crushing value (%)	Not measured	11.7	20.6

123

124 2.2 Mixture proportions

125 Eight mixtures were prepared in this study, as presented in Table 3. The water-to-cementitious
 126 materials ratio (W/C+F) and the water content were fixed at 0.30 and 165 kg/m³, respectively. The
 127 replacement ratios of cement with FA were 0%, 20%, and 40% by mass, and those of coarse
 128 aggregate with PCWA were 0% (G0) and 10% (G10) by volume. For the concretes including 20
 129 mass% and 40 mass% of FA in the cementitious materials, 20 vol.% (G20) of the coarse aggregate
 130 was also replaced with PCWA. The grading of the PCWA itself as well as the mixed coarse
 131 aggregates are presented in Fig. 1. The range limits of the 5–20 mm coarse aggregate defined as
 132 “Crushed stone 2005 for concrete” as per JIS A 5005 [35] are also shown in Fig. 1. Although the
 133 PCWA was finer than the reference coarse aggregate (G0), the differences in grading among G0,
 134 G10, and G20 were negligible within the low replacement ratios. A superplasticizer and an
 135 air-entraining agent were also used to obtain the same slump of 20.0 ± 2.0 cm and air content of 4.5
 136 ± 1.0%. The slump, air content, and temperature of the concrete, measured just after mixing, are
 137 also listed in Table 3.

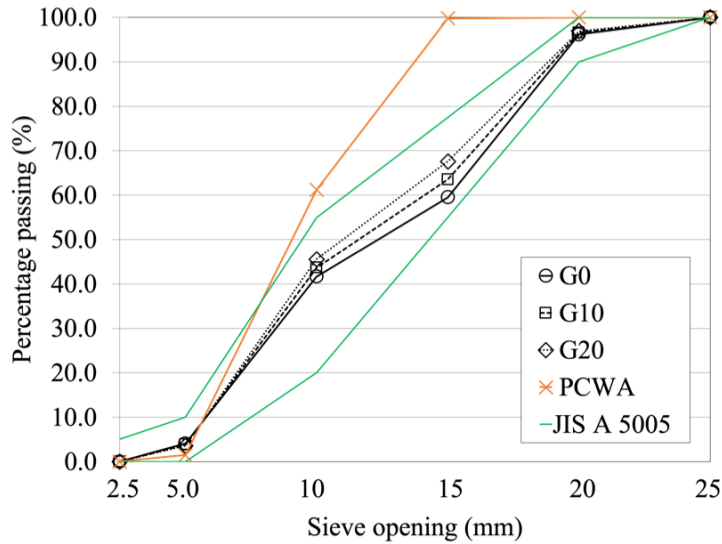
138

139 Table 3 Mixture proportions of concrete and measured properties of fresh concrete

	Unit content (kg/m ³)						Properties of fresh concrete		
	W	C	F	S	G	PCWA	Slump (cm)	Air content (%)	Temperature (°C)
HF0-G0	165	550	0	751	854	0	19.0	3.9	21.0
HF0-G10	165	550	0	751	780	74	20.0	3.9	19.0
HF20-G0	165	440	110	714	854	0	18.5	3.5	19.0
HF20-G10	165	440	110	714	780	74	20.0	3.8	20.0
HF20-G20	165	440	110	714	684	147	21.3	4.0	20.5

HF40-G0	165	330	220	677	854	0	20.0	3.8	19.0
HF40-G10	165	330	220	677	780	74	21.5	3.5	18.5
HF40-G20	165	330	220	677	684	147	21.3	4.2	20.1

140



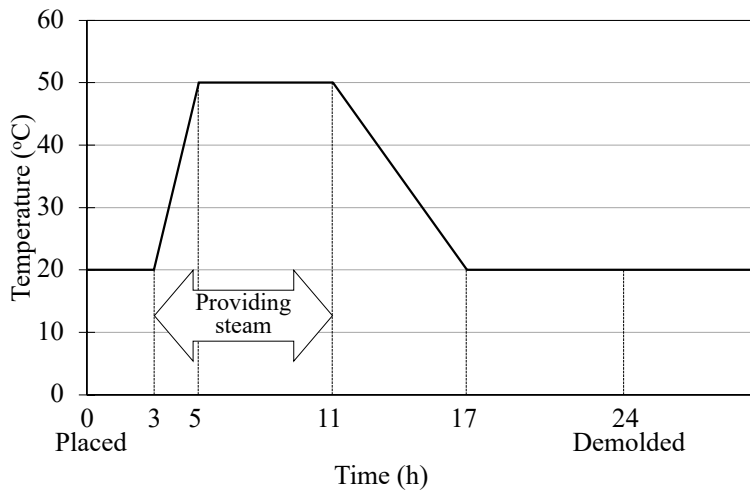
141

142 Figure 1 Gradings of coarse aggregates

143

144 2.3 Curing conditions

145 All the concrete specimens were cured under a steam curing condition after casting. One of the
 146 general steam curing conditions, as described in Fig. 2, was applied. The temperature was raised to
 147 50 °C at a rate of 15 °C/h after initial curing for 3 h at 20 °C and 60% relative humidity (RH).
 148 Thereafter, it was maintained at the maximum temperature of 50 °C for 6 h. The chamber was
 149 cooled down to 20 °C at a rate of 5 °C/h and then maintained at 20 °C for 7 h. Then, the specimens
 150 were demolded 24 h after casting and stored at 20 ± 2 °C and 60 ± 2% RH. The specimens were
 151 steam-cured from the age of 3 to 11 h during the time when the temperature was raised and
 152 maintained at the maximum.



153

154 Figure 2 Temperature history of steam curing

155

156 2.4 Specimens and testing procedure

157 2.4.1 Compressive strength

158 Compressive strength tests were conducted at the ages of 1, 7, 28, 56, 91, 182, and 364 d in
 159 accordance with JIS A 1108 (Method of test for compressive strength of concrete) [37]. Three
 160 cylindrical specimens, each measuring 100 mm in diameter and 200 mm in height, were used for
 161 each condition.

162

163 2.4.2 Length change

164 A prism specimen, 100 mm in height, 100 mm in width, and 400 mm in length, was prepared to
 165 measure the variation of length in the concrete. The change in length was measured with an
 166 embedded gauge (KM-100BT, Tokyo Measuring Instruments Lab., Japan) located at the center of
 167 the prismatic specimens immediately after placing. To evaluate the length change in the concrete
 168 due to autogenous shrinkage and drying shrinkage, the temperature in the specimen was also
 169 measured with a thermocouple included in the gauge to obtain the thermal strain from the measured

170 free strain. Similar to the measurements of the compressive strength of the specimens, these
171 specimens were also steam-cured and demolded at the age of 1 d. Then, they were stored at 20 °C
172 and 60% RH.

173

174 2.4.3 Accelerated carbonation

175 Three prism specimens, each measuring 100 mm in height, 100 mm in width, and 400 mm in length,
176 were prepared for the accelerated carbonation test for each mixture. These specimens were cured
177 under the aforementioned conditions until the age of 91 d, and were then stored at 20 ± 2 °C, $60 \pm$
178 5% RH, and $5.0 \pm 0.2\%$ CO₂. The age of 91 d was selected because FA concrete would have been
179 sufficiently cured within this time. All the surfaces, except for one of the 100 × 400 mm sides,
180 which was not the top or bottom surface at the casting stage, were coated with epoxy resin. Only the
181 uncoated 100 × 400 mm side was exposed to the accelerated carbonation condition. The
182 carbonation depths at six positions for each specimen were measured using the phenolphthalein
183 method at the accelerated carbonation periods of 1, 4, 8, 13, and 26 weeks.

184

185 2.4.4 Pore size distribution

186 The pore size distributions of six mixtures, namely HF0-G0, HF0-G10, HF20-G0, HF20-G10,
187 HF40-G0, and HF40-G10, were measured at the age of 364 d using a mercury intrusion porosimeter
188 (POREMASTER 60, Quantachrome Instruments, USA). The samples for the test, each ranging
189 from 2.5 to 5.0 mm in size, were obtained after the compressive strength test was completed. These
190 samples were soaked in acetone to stop further hydration, and then dried in a vacuum desiccator
191 before the test. The pore size distribution was measured from 3 nm to 300 μm.

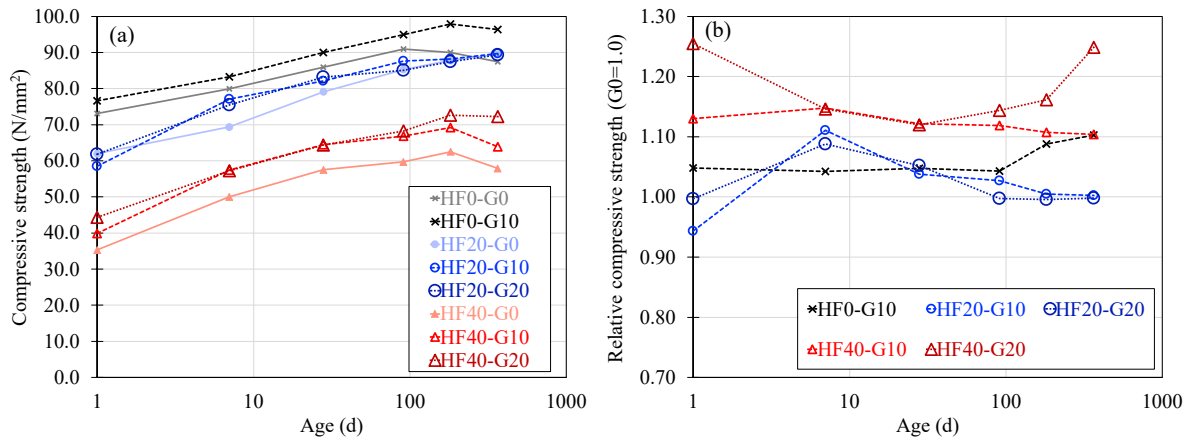
192

193 3 Results and discussion

194 3.1 Compressive strength

195 The compressive strengths of all the mixtures are shown in Fig. 3 (a), and the standard errors ranged
196 from 0.2 to 4.6 N/mm². In addition, the compressive strengths normalized by the strength of each
197 mixture without PCWA are shown in Fig. 3 (b). Although the compressive strength decreased with
198 the increase in FA fraction, the values for HF20-G0, HF20-G10, and HF20-G20 at the age of 364 d
199 were almost the same as that of HF0-G0 (see Fig. 3 (a)). It implies that when FA was used with the
200 low replacement ratio of 20%, it may have reacted well and contributed to the development of the
201 same compressive strength as that of the concrete using only high-early-strength Portland cement in
202 the long term. It can be found that the compressive strengths of concrete using PCWA were equal to
203 or higher than those without PCWA in all the mixtures and for all the ages except for HF20-G10 at
204 the age of 1 d. This indicates that the internal curing using PCWA is likely to have developed the
205 compressive strength of concrete. The PCWA with higher crushing value than conventional
206 aggregates has the risk of inducing strength reduction of concrete, while the reduction rate should
207 depend on the replacement ratio. According to a previous study [27], however, the compressive
208 strength of ultra-high-strength concrete with the replacement of 40% coarse aggregate by PCWA
209 was 160 N/mm², which was 30% higher than the ultra-high-strength concrete without PCWA. That
210 is, there were no particular negative effects of PCWA on the strength development of concrete.
211 Moreover, judging from the present test results, the strength reduction is not significant so long as
212 the replacement ratio is below 20% and the enhancement effect of the internal curing is high beyond
213 this negative effect. According to Fig. 3(b), using PCWA can improve the compressive strength of
214 concrete by up to 25%. This means that the internal water of PCWA can be supplied to the mortar
215 matrix naturally, and the PCWA can cure the concrete internally. In the case of concrete using only
216 high-early-strength Portland cement, the gain in compressive strength using 10% PCWA was
217 approximately 5% at the age of 1 d and increased to 10% at the ages of 182 and 364 d (see Fig. 3

218 (b)). This means that the effect of PCWA increased slightly over time. However, when the FA
219 replacement ratio was 20%, the PCWA increased the compressive strength by 10% and 5% at the
220 ages of 7 and 28 d, respectively, and the compressive strength of concrete using PCWA was almost
221 the same as that without PCWA at the other ages regardless of the PCWA replacement ratio. It is
222 assumed that the internal water of PCWA could participate in the development of compressive
223 strength at a relatively early age. In the case of the 40% FA replacement, the gain in compressive
224 strength due to PCWA was more than 10% for the early and later ages. The normalized
225 compressive strength of HF40-G20 was 1.25 at the ages of 1 and 364 d, which indicated that the
226 PCWA increased the compressive strength by 25%. Notably, this was the highest value for all the
227 ages. A previous study reported that the application of 40% PCWA to high-strength FA concrete
228 using 60% ordinary Portland cement and 40% FA with the water-to-cementitious materials ratio of
229 0.30 improved the compressive strength by 8.4–16.5% when it was cured under sealed conditions at
230 20 °C [29]. When compared to the result from the previous study [29], the addition of 20% PCWA
231 improved the compressive strength to quite an extent both at the early and later ages, whereas while
232 the addition of 10% PCWA provided a notable improvement in the compressive strength at the
233 early age, this gain gradually decreased. This implies that the use of PCWA at the replacement ratio
234 of 20% in the case of the 40% FA fraction in steam-cured concrete could effectively enhance
235 cement hydration at an early age and the FA reaction in the long term. Furthermore, when compared
236 with the previous study [29], the development of the compressive strength of the FA concrete in this
237 study was not significant even in the long term, and the compressive strengths of HF40-G10 and
238 HF40-G20 decreased slightly from 182 to 364 d. This result may be attributed to the negative
239 impact of the steam curing, that is, the acceleration of cement hydration and FA reaction at the early
240 age.



241

242 Figure 3 Effect of PCWA on compressive strength: (a) compressive strength of concrete with age,

243 (b) compressive strength normalized by that of concrete without PCWA

244 3.2 Length change

245 The length change until the age of 1 d, as shown in Fig. 4, was obtained by subtracting the thermal

246 strain from the measured free strain, where the thermal expansion coefficient is assumed to be a

247 constant of $10 \times 10^{-6}/^{\circ}\text{C}$. It should be noted that the first peak of the measured value of length

248 change in concrete corresponded to zero shrinkage in concrete up to the age of 1 d, as seen in Fig. 4.

249 For the FA replacements, except for HF20-G10, the shrinkage in concrete using PCWA was smaller

250 than that without PCWA. This result implies that PCWA can effectively reduce the autogenous

251 shrinkage by supplying the internal water to the mortar matrix self-desiccated due to hydration. For

252 the FA concrete, a PCWA replacement ratio of 20% could reduce the autogenous shrinkage to a

253 greater extent than the replacement ratio of 10%. The shrinkage in Fig. 4, however, could be

254 overestimated with the assumption that the thermal expansion coefficient is $10 \times 10^{-6}/^{\circ}\text{C}$ as this

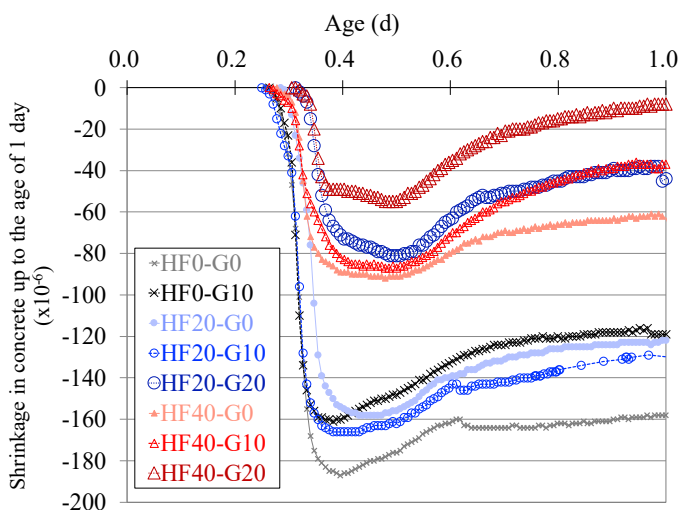
255 value can change at early ages [38]. Thus, further investigation into the autogenous shrinkage while

256 considering the time dependency of the thermal expansion coefficient is required to ascertain its

257 absolute value.

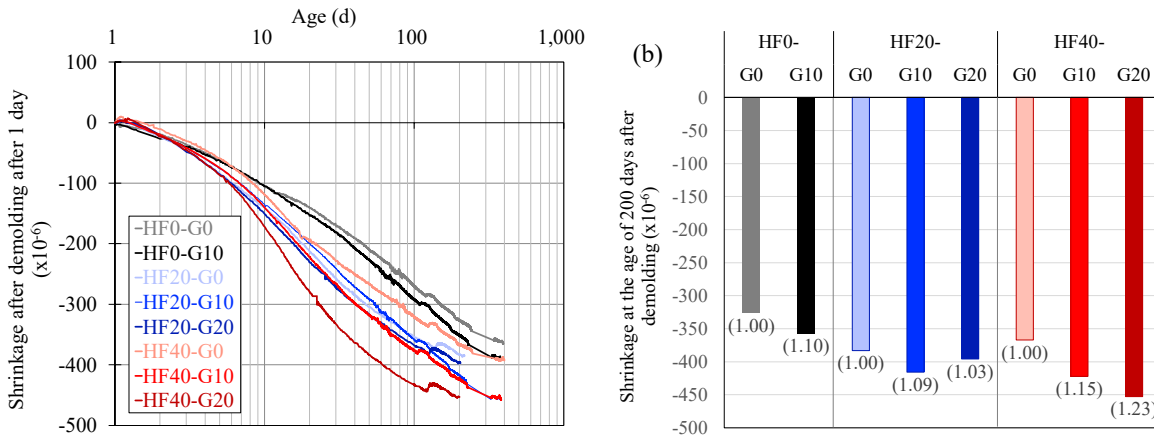
258 The length changes immediately after demolding and at the age of 200 d after demolding are also

259 shown in Fig. 5 (a) and (b), respectively. The value of zero length change was set to the strain at the
 260 age of 1 d (i.e., at the time of the demolding). Although there was no significant difference in the
 261 length change until the age of 3 d, the addition of PCWA increased the length change in concrete
 262 after its exposure to air until the age of 200 d, especially for the concrete with 40% FA (see Fig. 5
 263 (b)). It appears that the internal water supplied during steam curing may not have been consumed
 264 completely for cement hydration and evaporated at a later age, resulting in the higher drying
 265 shrinkage in the concrete with PCWA at a higher replacement ratio, especially when the
 266 water-to-cement ratio was relatively high (i.e., the FA replacement ratio was high). Although the
 267 internal water of PCWA could play a role in internal curing, the water supplied from PCWA also
 268 could evaporate and cause drying shrinkage. This idea is in line with the results of a previous study
 269 [30]; though the length change in concrete without PCWA was almost the same or lower than that
 270 in concrete with PCWA for the same mass loss in concrete after demolding, the mass loss in
 271 concrete with PCWA increased to a greater extent than that in the concrete without PCWA,
 272 resulting in the larger shrinkage in the former.



273

274 Figure 4 Shrinkage in concrete up to the age of 1 d before demolding



275

276 Figure 5 Shrinkage in concrete after demolding at 1 d: (a) with age, (b) at the age of 200 d

277

278 3.3 Accelerated carbonation

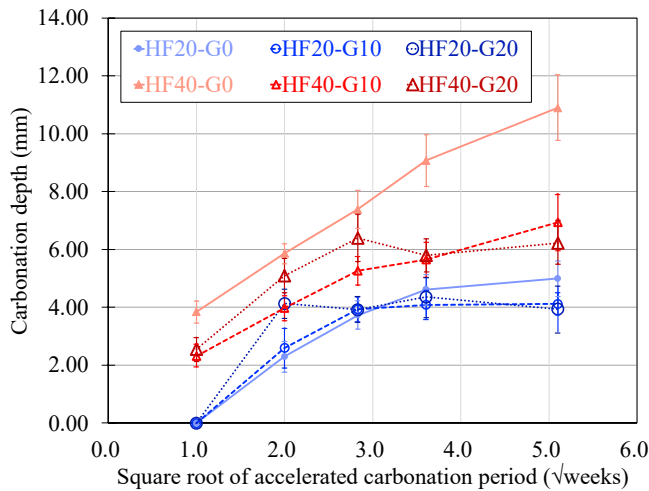
279 The carbonation depths of FA concrete are shown in Fig. 6. It can be observed that the carbonation
 280 depth of HF40-G0 was higher than that of any other mixture regardless of the carbonation period.

281 Thus, the internal curing by PCWA was effective in lowering the carbonation depth for the concrete
 282 using 40% FA, whereas no significant effect of PCWA was observed for the concrete using 20%

283 FA. The tendency was the same as that of compressive strength (see Fig. 3 (a)). Moreover, the
 284 carbonation rate after 4 or 8 weeks reduced compared to that before 4 or 8 weeks, except for

285 HF40-G0. It implies that the steam curing and storage at 60% RH made the surface layer of the
 286 concrete porous, resulting in fast carbonation, whereas long-term storage had relatively less

287 influence within the concrete, resulting in slow carbonation. Further investigation into properties
 288 such as pore structure is needed to confirm the aforementioned idea of inhomogeneity.



289

290 Figure 6 Carbonation depth of FA concrete under accelerated carbonation conditions

291

292 The carbonation coefficients were calculated using regression analysis with an empirical
 293 relationship based on the square root law, as expressed by Eq. (1). The carbonation coefficients
 294 were obtained using all the plotted data in Fig. 6.

295
$$y = \alpha\sqrt{t} \quad (1)$$

296 where y is the carbonation depth (mm), t is the period of accelerated carbonation (week), and α is
 297 the accelerated carbonation coefficient ($\text{mm}/\text{week}^{0.5}$). As shown in Table 4, the accelerated
 298 carbonation coefficients of HF40-G10 and HF40-G20 were approximately 65% of that of HF40-G0.
 299 It appears that internal curing by the PCWA could have effectively enhanced the carbonation
 300 resistance in concrete using 40% FA by approximately 35%. This means that the internal water of
 301 PCWA can be supplied to the mortar matrix as curing water to promote cement hydration and FA
 302 reaction, resulting in denser microstructure and enhancement of carbonation resistance. The
 303 relationships between the accelerated carbonation coefficient and compressive strength at each age
 304 are shown in Fig. 7. The compressive strength increased with the decrease in the accelerated
 305 carbonation coefficient regardless of the inclusion of PCWA. The effects of the internal curing on

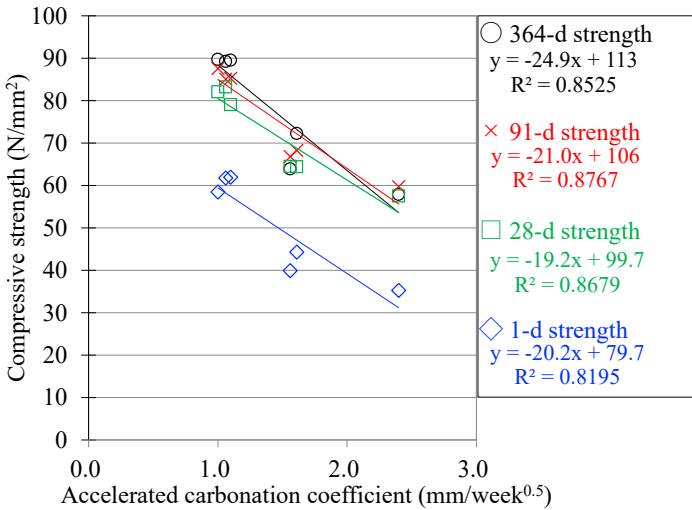
306 compressive strength as well as carbonation resistance could be attributed to the improvement in
 307 pore structure. That is, using PCWA can make the concrete microstructure denser, resulting in
 308 better quality concrete, as reiterated in the next section. Furthermore, the relationship between the
 309 accelerated carbonation coefficient and compressive strength suggests that the CO₂ gas might have
 310 penetrated only the mortar matrix, and that the porous structure of PCWA did not adversely affect
 311 the carbonation resistance of concrete since the relationship can be approximated by a line
 312 regardless of the PCWA replacement. Further, the carbonation rate can be estimated using the
 313 compressive strength even when adding PCWA.

314

315 Table 4 Accelerated carbonation coefficient (mm/week^{0.5})

HF20-G0	HF20-G10	HF20-G20	HF40-G0	HF40-G10	HF40-G20
1.10	1.00	1.06	2.40	1.56	1.60

316



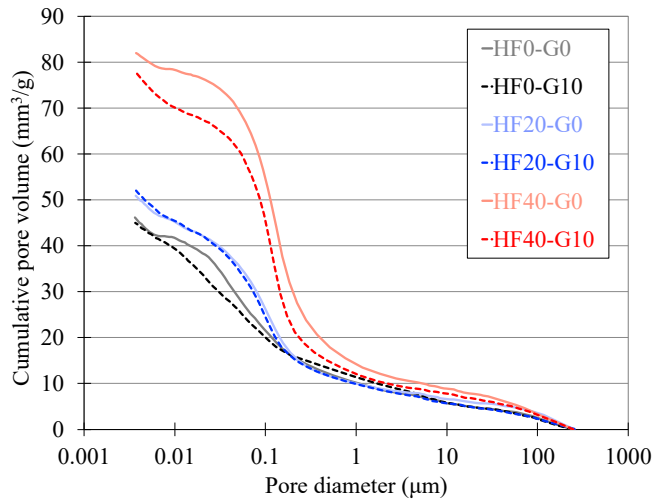
317

318 Figure 7 Relationship between compressive strength and accelerated carbonation coefficient

319

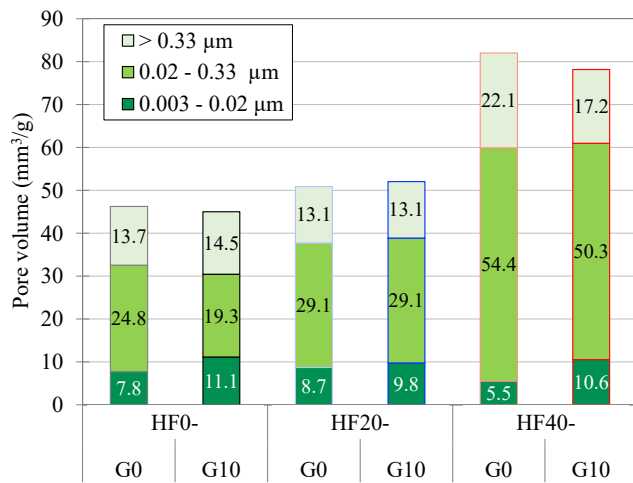
320 3.4 Pore size distribution

321 Figure 8 shows the cumulative intrusion curves for six samples at the age of 364 d [32]. Although
322 using 10% PCWA in the concrete with 40% FA replacement slightly reduced the cumulative pore
323 volume, this effect was not observed clearly for the concrete with 0% and 20% FA replacements.
324 The trend was almost the same as that of the effects on compressive strength and carbonation
325 resistance. Figure 9 shows the pore size distributions of these samples. According to Yamamoto and
326 Kanazu [39], the pore volume for the pore size ranges of 0.003 μm to 0.02 μm increased, whereas
327 that for the size ranges of 0.02 μm to 0.33 μm decreased as the pozzolanic reaction of FA
328 progressed. Figure 9 also shows that the volumes of pores smaller than 0.02 μm increased
329 significantly and those of pores ranging from 0.02 μm to 0.33 μm in size decreased for HF40
330 concrete with PCWA. This result indicates that the internal curing using PCWA could effectively
331 enhance the pozzolanic reaction of FA in HF40 concrete, resulting in higher compressive strength
332 and a lower accelerated carbonation coefficient. Moreover, the pore size distribution of HF20-G10
333 was almost the same as that of HF20-G0, which reflected the slight effect of PCWA on the
334 compressive strength and carbonation resistance of steam-cured concrete using 20% FA
335 replacement. The same pore size distribution tendency as that of HF40 concrete can be observed for
336 the HF0 concrete (i.e., in the concrete without FA). This result may also indicate the promotion of
337 cement hydration by PCWA, which is in agreement with the findings of a previous study [29].
338 These results imply that the internal water of PCWA can enhance the cement hydration and FA
339 reaction by being supplied as the curing water. Moreover, a denser pore structure can be formed.



340

341 Figure 8 Cumulative intrusion curves of the concrete specimens at the age of 364 d [32]



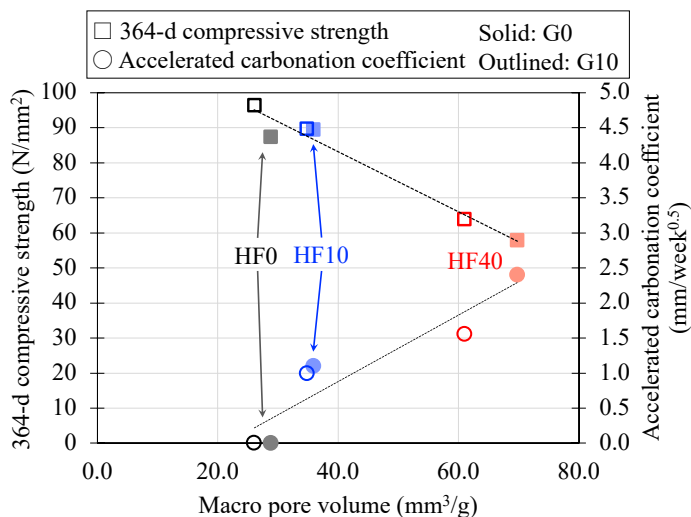
342

343 Figure 9 Pore size distributions of the concrete specimens at the age of 364 d

344

345 In general, macropores (pores larger than 0.05 µm) play a dominant role in the mechanical
 346 properties and mass transfer resistance of concrete [40]. Figure 10 shows the relationship between
 347 the macropore volume and the 364-d compressive strength. The relationships between the
 348 macropore volume and the accelerated carbonation coefficient are also shown in Fig. 10, wherein
 349 the accelerated carbonation coefficient of HF0 is plotted as 0. As the macropore volume reduces,
 350 the compressive strength increases and the accelerated carbonation coefficient reduces regardless of

351 the mixture proportion. It can also be observed that for HF40 concrete alone, the use of PCWA
 352 decreased the macropore volume, resulting in higher compressive strength and lower accelerated
 353 carbonation coefficient. Consequently, internal curing using PCWA can assist in generating a
 354 denser microstructure and improving the compressive strength and carbonation resistance.



355

356 Figure 10 Relationship between macropore volume and other properties

357

358 4. Conclusions

359 The purpose of this study was to investigate the effects of PCWA as an internal curing agent on the
 360 compressive strength, shrinkage, and carbonation of steam-cured FA concrete, where the
 361 replacement ratios of FA were 0%, 20%, and 40% by mass, and those of PCWA were 0%, 10%,
 362 and 20% by volume.

363 The following conclusions can be drawn:

- 364 (1) The internal curing using PCWA effectively enhanced the compressive strength of steam-cured
 365 FA concrete, especially when the replacement ratio of FA was 40%. PCWA increased the
 366 compressive strength of the concrete using 40% FA by 10–25%. This effect was obtained at the
 367 early age as well as in the long term. No adverse effect owing to the porous structure of PCWA

368 was observed for up to 20% replacement by PCWA up to the age of 364 d.

369 (2) PCWA could effectively reduce autogenous shrinkage and increase drying shrinkage in the
370 steam-cured FA concrete.

371 (3) The accelerated carbonation resistance in the steam-cured concrete with 40% FA improved
372 significantly with the addition of PCWA, and the effects of PCWA at the replacement ratios of
373 10% and 20% on the carbonation resistance were almost the same. PCWA replacements of 10%
374 and 20% reduced the accelerated carbonation coefficient of 40% FA concrete by approximately
375 35%.

376 (4) The inverse linear relationship between the compressive strength and the accelerated
377 carbonation coefficient was observed regardless of PCWA replacement. This result clarified
378 that the CO₂ mainly penetrates the mortar matrix, not the PCWA, and that the porous structure
379 of PCWA does not affect the carbonation resistance of concrete adversely.

380 (5) Considering the pore size distribution, the internal curing using PCWA could effectively
381 enhance the FA reaction and cement hydration. PCWA decreased the macropore volume in
382 mortar matrix, resulting in enhanced compressive strength and improved carbonation resistance.

383 Consequently, although PCWA is derived from waste, it is beneficial as an internal curing agent to
384 improve the quality of not only steam-cured cement concrete but also steam-cured FA concrete. The
385 quality improvement from using PCWA can greatly contribute to promoting the utilization of the
386 waste of PCWA as well as the by-products of FA in the construction field. Further investigations on
387 microstructure and/or internal relative humidity change of the concrete with time are needed to
388 explain the mechanism of the internal curing effect of PCWA. This would also be useful for the
389 design of the PCWA replacement ratio.

390

391 Conflicts of interests

392 The authors declare no conflicts of interest at this time.

393

394 Acknowledgment

395 The authors would like to thank Mr. Naoki Doi and Mr. Yusuke Muragishi, Master's students at
396 Hiroshima University, for their help in conducting the experiments. This research was partially
397 supported by a JSPS KAKENHI Grant-in-Aid for Young Scientists (B) (No. JP25820191).

398

399 References

- 400 [1] J. Bijen, Benefits of slag and fly ash, *Constr. Build. Mater.* 10 (1996) 309–314.
401 [https://doi.org/10.1016/0950-0618\(95\)00014-3](https://doi.org/10.1016/0950-0618(95)00014-3).
- 402 [2] T.R. Naik, B.W. Ramme, High early strength fly ash concrete for precast/prestressed
403 products, *PCI J.* 35 (1990) 72–78. <https://doi.org/10.15554/pcij.11011990.72.78>.
- 404 [3] J. Zachar, Sustainable and economical precast and prestressed concrete using fly ash as a
405 cement replacement, *J. Mater. Civ. Eng.* 23 (2011) 789–792.
406 [https://doi.org/10.1061/\(asce\)mt.1943-5533.0000243](https://doi.org/10.1061/(asce)mt.1943-5533.0000243).
- 407 [4] Z. He, J. Liu, K. Zhu, Influence of mineral admixtures on the short and long-term
408 performance of steam-cured concrete, *Energy Procedia* 16 (2011) 836–841.
409 <https://doi.org/10.1016/j.egypro.2012.01.134>.
- 410 [5] B. Liu, Y. Xie, J. Li, Influence of steam curing on the compressive strength of concrete
411 containing supplementary cementing materials, *Cem. Concr. Res.* 35 (2005) 994–998.
412 <https://doi.org/10.1016/j.cemconres.2004.05.044>.
- 413 [6] B. Lothenbach, F. Winnefeld, C. Alder, E. Wieland, P. Lunk, Effect of temperature on the
414 pore solution, microstructure and hydration products of Portland cement pastes, *Cem.*
415 *Concr. Res.* 37 (2007) 483–491. <https://doi.org/10.1016/j.cemconres.2006.11.016>.

- 416 [7] D.W.S. Ho, C.W. Chua, C.T. Tam, Steam-cured concrete incorporating mineral admixtures,
417 Cem. Concr. Res. 33 (2003) 595–601. [https://doi.org/10.1016/S0008-8846\(02\)01028-1](https://doi.org/10.1016/S0008-8846(02)01028-1).
- 418 [8] G. Long, Z. He, A. Omran, Heat damage of steam curing on the surface layer of concrete,
419 Mag. Concr. Res. 64 (2012) 995–1004. <https://doi.org/10.1680/mac.11.00164>.
- 420 [9] S. Nie, S. Hu, F. Wang, P. Yuan, Y. Zhu, J. Ye, Y. Liu, Internal curing – A suitable method
421 for improving the performance of heat-cured concrete, Constr. Build. Mater. 122 (2016)
422 294–301. <https://doi.org/10.1016/j.conbuildmat.2016.05.159>.
- 423 [10] C. Zou, G. Long, C. Ma, Y. Xie, Effect of subsequent curing on surface permeability and
424 compressive strength of steam-cured concrete, Constr. Build. Mater. 188 (2018) 424–432.
425 <https://doi.org/10.1016/j.conbuildmat.2018.08.076>.
- 426 [11] P. Lura, Autogenous Deformation and Internal Curing of Concrete, Delft University Press,
427 Delft, 2003.
- 428 [12] CT-18: ACI Concrete Terminology, American Concrete Institute, Farmington Hills, Mich.,
429 (2018) 36.
- 430 [13] R. Philleo, Concrete science and reality, in: J.P. Skalny, S. Mindess (Eds.), Materials
431 Science of Concrete II, American Ceramic Society, Westerville, Ohio, 1991, pp. 1–8.
- 432 [14] T.A. Hammer, High strength LWA concrete with silica fume effect of water content in the
433 LWA on mechanical properties, in: Suppl. Pap. Fourth CANMET/ACI Int. Conf. Fly Ash,
434 Silica Fume, Slag, Nat. Pozzolans Concr., Istanbul, Turkey, 1992: pp. 314–330.
- 435 [15] D.P. Bentz, P. Lura, J.W. Roberts, Mixture proportioning for internal curing, Concr. Int. 27
436 (2005) 35–40.
- 437 [16] D.P. Bentz, Internal curing of high-performance blended cement mortars, ACI Mater. J. 104
438 (2007) 408–414.
- 439 [17] D. Cusson, T. Hoogeveen, Internal curing of high-performance concrete with pre-soaked

- 440 fine lightweight aggregate for prevention of autogenous shrinkage cracking, *Cem. Concr.*
441 *Res.* 38 (2008) 757–765. <https://doi.org/10.1016/j.cemconres.2008.02.001>.
- 442 [18] O.M. Jensen, P.F. Hansen, Water-entrained cement-based materials, *Cem. Concr. Res.* 31
443 (2001) 647–654. [https://doi.org/10.1016/s0008-8846\(01\)00463-x](https://doi.org/10.1016/s0008-8846(01)00463-x).
- 444 [19] B. Craeye, M. Geirnaert, G. De Schutter, Super absorbing polymers as an internal curing
445 agent for mitigation of early-age cracking of high-performance concrete bridge decks,
446 *Constr. Build. Mater.* 25 (2011) 1–13. <https://doi.org/10.1016/j.conbuildmat.2010.06.063>.
- 447 [20] J. Justs, M. Wyrzykowski, D. Bajare, P. Lura, Internal curing by superabsorbent polymers
448 in ultra-high performance concrete, *Cem. Concr. Res.* 76 (2015) 82–90.
449 <https://doi.org/10.1016/j.cemconres.2015.05.005>.
- 450 [21] O.M. Jensen, P. Lura, Techniques and materials for internal water curing of concrete, *Mater.*
451 *Struct. Constr.* 39 (2006) 817–825. <https://doi.org/10.1617/s11527-006-9136-6>.
- 452 [22] S.T. Yildirim, C. Meyer, S. Herfellner, Effects of internal curing on the strength, drying
453 shrinkage and freeze-thaw resistance of concrete containing recycled concrete aggregates,
454 *Constr. Build. Mater.* 91 (2015) 288–296.
455 <https://doi.org/10.1016/j.conbuildmat.2015.05.045>.
- 456 [23] F. Liu, J. Wang, X. Qian, J. Hollingsworth, Internal curing of high performance concrete
457 using cenospheres, *Cem. Concr. Res.* 95 (2017) 39–46.
458 <https://doi.org/10.1016/j.cemconres.2017.02.023>.
- 459 [24] M. Wyrzykowski, S. Ghourchian, S. Sinthupinyo, N. Chitvoranund, T. Chintana, P. Lura,
460 Internal curing of high performance mortars with bottom ash, *Cem. Concr. Compos.* 71
461 (2016) 1–9. <https://doi.org/10.1016/j.cemconcomp.2016.04.009>.
- 462 [25] M. Suzuki, I. Maruyama, T. Kawabata, R. Sato, A study on deformation of ultra high
463 strength concrete containing crushed roof tile aggregate (in Japanese), *Proc. Japan Concr.*

- 464 Inst. 29 (2007) 651–656.
- 465 [26] S. Suzuki, Survey research on manufacture and supply of structural concrete using crushed
466 roof tile waste as aggregate (in Japanese), *Japan Test. Cent. Constr. Mater. J.* 51 (2015)
467 14–19.
- 468 [27] M. Suzuki, M. Seddik Meddah, R. Sato, Use of porous ceramic waste aggregates for
469 internal curing of high-performance concrete, *Cem. Concr. Res.* 39 (2009) 373–381.
470 <https://doi.org/10.1016/j.cemconres.2009.01.007>.
- 471 [28] R. Sato, A. Shigematsu, T. Nukushina, M. Kimura, Improvement of properties of Portland
472 blast furnace cement type B concrete by internal curing using ceramic roof material Waste,
473 *J. Mater. Civ. Eng.* 23 (2010) 777–782.
474 [https://doi.org/10.1061/\(asce\)mt.1943-5533.0000232](https://doi.org/10.1061/(asce)mt.1943-5533.0000232).
- 475 [29] P.T. Bui, Y. Ogawa, K. Nakarai, K. Kawai, R. Sato, Internal curing of Class-F fly-ash
476 concrete using high-volume roof-tile waste aggregate, *Mater. Struct.* 50 (2017) 203.
477 <https://doi.org/10.1617/s11527-017-1073-z>.
- 478 [30] Y. Muragishi, Y. Ogawa, K. Kawai, R. Sato, Effect of porous ceramic waste aggregate on
479 durability of steam cured fly ash concrete (in Japanese), *Cem. Sci. Concr. Technol.* 68
480 (2014) 337–344. <https://doi.org/10.14250/cement.68.337>.
- 481 [31] P.T. Bui, Y. Ogawa, N. Doi, K. Kawai, R. Sato, Properties of steam-cured fly ash concrete
482 using porous ceramic waste aggregate, in: *Proc. 13th Int. Conf. Recent Adv. Concr.*
483 *Technol. Sustainability Issues*, Ottawa, Canada, ACI SP303, 2015: pp. 323–336.
- 484 [32] P.T. Bui, Y. Muragishi, Y. Ogawa, K. Kawai, R. Sato, Effects of porous ceramic waste
485 aggregate as an internal curing agent on steam-cured high strength fly ash concrete, in: *Proc.*
486 *Int. Conf. Sustainable Struct. Concr.*, La Plata, Argentina, 2015: pp. 66–76.
- 487 [33] Japanese Standards Association, JIS R 5210:2009. Portland cement, Japanese Standards

- 488 Association, Tokyo, Japan, 2009.
- 489 [34] Japanese Standards Association, JIS R 6201:2015. Fly ash for use in concrete, Japanese
490 Standards Association, Tokyo, Japan, 2015.
- 491 [35] Japanese Standards Association, JIS A 5005:2009. Crushed stone and manufactured sand
492 for concrete, Japanese Standards Association, Tokyo, Japan, 2009.
- 493 [36] British Standards Institution, BS 812-110:1990. Testing aggregates. Methods for
494 determination of aggregate crushing value (ACV), British Standards Institution, London,
495 United Kingdom, 1990.
- 496 [37] Japanese Standards Association, JIS A 1108:2018. Method of test for compressive strength
497 of concrete, Japanese Standards Association, Tokyo, Japan, 2018.
- 498 [38] I. Maruyama, A. Teramoto, Impact of time-dependant thermal expansion coefficient on the
499 early-age changes in cement paste, *Cem. Concr. Res.* 41 (2011) 380–391.
500 <https://doi.org/10.1016/j.cemconres.2011.01.003>.
- 501 [39] T. Yamamoto, T. Kanazu, Experimental explanation of compacting effect on hydration
502 phases and strength development mechanism derived from pozzolanic reaction of fly ash
503 (in Japanese), *J. Japan Soc. Civ. Eng. Ser. E.* 63 (2007) 52–65.
- 504 [40] P.K. Mehta, P.J. Monteiro, *Concrete. Microstructure, properties, and materials*, third ed.,
505 McGraw-Hill, New York, 2006.

506