

1 Impact of Aggregate Properties on the Development of Shrinkage-Induced
2 Cracking in Concrete under Restraint Conditions

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18 **ABSTRACT**

19 Concrete specimens were prepared with the same mixture proportion except for their
20 constituent coarse aggregates, namely limestone and sandstone that possess different inherent
21 drying shrinkage values. The strain at a cross section perpendicular to the drying direction
22 under restricted and unrestrained conditions was observed using a digital image correlation
23 method. It was confirmed that initiation and propagation of cracks were greatly affected by
24 the types of aggregate. Analysis by the rigid-body spring network method, which explicitly
25 incorporates aggregate shrinkage and properties of the interfacial transition zone (ITZ),
26 revealed that aggregate shrinkage and fracture energy of the ITZ greatly influence fine-crack
27 distribution and localization of cracks. It was elucidated that pure limestone with a small
28 drying shrinkage value can reduce the number of visible cracks in concrete under restraint
29 conditions since it allows fine cracks to form around coarse aggregate particles that absorb
30 the localization of cracks, thus limiting wider cracks.

31

32 **Keywords:** Aggregate (D); Interfacial Transition Zone (B); Drying (A); Microcracking (B);
33 Shrinkage (C).

34

35

36 1. Introduction

37 Cracking in concrete produced by drying shrinkage is an undesirable phenomenon since it
38 causes disfigurement of building surfaces and jeopardizes the durability of concrete by
39 facilitating the ingress of water, carbon dioxide, and other aggressive substances. Hence, the
40 shrinkage mechanism of cement paste [1-10], aggregate properties and their restraint
41 performances in concrete [11-20], the effect of aggregate particle size on shrinkage [11,
42 21-23], water movement behavior [17, 24-32], and formulae to predict drying shrinkage of
43 concrete based on mixture proportions and environmental conditions [33, 34] have been
44 thoroughly studied. Due to the high volume ratio of aggregates in concrete, the impact of
45 aggregate properties on concrete shrinkage has gained fresh attention in recent research,
46 consequently leading to more intense investigations.

47

48 Meanwhile, the behavior of concrete under restraint conditions has not been characterized as
49 well as its behavior under free conditions. While numerous studies on cracking behavior and
50 criteria for the initiation of through-cracking criteria have been conducted [35-40], in general,
51 restrained concrete shows a complex behavior during the drying process. Concrete members
52 contain a gradation of water content, which results in intrinsic shrinkage strains that are
53 mutually restrained by each other within each concrete member and by an outer restraining
54 body. Considering this inhomogeneity of stress in concrete members, the majority of the
55 research has focused on engineering or design purposes but not on better understanding of
56 material characteristics. Thus, in addition to through-cracking, the initiation of cracking and
57 the propagation process at the meso-scale (scale from micrometer to millimeter) should be
58 studied.

59

60 It has been reported that a series of concretes that had the same volumetric mixture

61 proportions except for coarse aggregate type showed different numbers of through-cracks in
62 reinforced concrete prism specimens under restraint conditions while differing in minor
63 surface cracking [41, 42]. Fig. 1 presents a comparison of surface crack patterns in concrete
64 with differing coarse aggregates. The crack patterns of two types of concrete recorded at 20°C
65 and a relative humidity (RH) of 60% are shown on the left of Fig. 1. These concrete types
66 contained a coarse aggregate of pure limestone and exhibited a concrete shrinkage of about
67 600 microns, while the concrete on the right side of Fig. 1 contained a coarse aggregate of
68 sandstone with a large amount of clay minerals and displayed a concrete shrinkage of 1000
69 microns. Concrete with limestone shows a smaller number of through-cracks than concrete
70 with sandstone as the aggregate.

71

72 The main driver of concrete shrinkage is the cement paste. A similar amount of cement paste
73 is used to make concrete with limestone and sandstone aggregates. Therefore, ignoring the
74 role of the cement paste allows an isolated comparison of the shrinkage caused purely by the
75 aggregates. Under restraint conditions, concrete members produce internal stresses, and
76 while the expected stresses in concretes with different aggregates are similar, their cracking
77 behaviors are quite different. While a concrete mix with a limestone aggregate had a small
78 number of through-cracks and a large number of minor surface cracks, concrete with
79 sandstone aggregates exhibited the opposite behavior [43]. In other words, the shrunk volume
80 of concrete is differently apportioned to through-cracks and minor/micro cracks depending
81 on the aggregate used.

82

83 The cracking behavior of concrete is generally considered to be highly complex. Studying
84 aggregate-dependent crack initiation can lead to a better understanding of this phenomenon
85 and aid in formulating mechanisms to control cracking.

86

87 Although crack propagation due to drying is difficult to observe experimentally, we recently
88 developed a measurement method for crack propagation using a digital image correlation
89 method (DICM) to overcome this issue [44]. In the present study, we first applied a
90 waterproof coating on the surface of concrete specimens to determine crack proportions in
91 fabricated concrete walls, as well as to provide the background color for the DICM analysis.
92 Second, as comparing numerical simulations with experimental data is quite informative [18,
93 19, 45, 46], we applied rigid-body-spring networks (RBSN) [47-49] to reproduce trends of
94 crack initiation and propagation behavior in order to understand the impact of aggregate
95 properties on these processes.

96

97 2. Experimental Techniques

98 2.1 Materials

99 Two concrete specimens with different shrinkage properties were prepared with a
100 water-to-cement ratio of 0.55 using ordinary Portland cement whose properties are
101 summarized in Table 1. The differences in shrinkage properties were realized by using two
102 different coarse aggregates, namely limestone (GL) and sandstone (GS). GL is very pure
103 limestone and shows almost no shrinkage, whereas GS has a large amount of chlorite and
104 shows large shrinkage. Short-term length change isotherms of GL and GS in three orthogonal
105 directions [50] are reproduced in Fig. 2. We denote concrete containing GL and GS as LS and
106 SS in this study, respectively. Concrete mixture proportions of LS and SS were designed to
107 keep the unit volume of coarse aggregate constant in order to isolate the effect of aggregate
108 properties on the total shrinkage. In addition, in order to eliminate size effects due to
109 differences in particle size distribution, each aggregate was first screened with sieves of 5 mm
110 to 10 mm, 10 mm to 15 mm, and 15 mm to 20 mm, and then the three grades of aggregates

111 were mixed uniformly. It should be noted that all the aggregates are prepared in saturated
112 surface dry conditions before mixing. Viscosity improver was used to avoid bleeding and
113 evade any changes in concrete quality; hence, fluidity was not controlled in this mix design.
114 Details of the materials used are listed in Table 2. The mix proportions of concrete and their
115 respective fresh properties are listed in Table 3 and the properties of the aggregates are listed
116 in Table 4. The details of the experiments carried out to study aggregate properties are
117 introduced in Section 2.2. Note that concrete specimens were demolded at the age of one day
118 and subjected to underwater curing using a saturated calcium hydroxide solution at a
119 temperature of $20^{\circ}\text{C} \pm 2^{\circ}\text{C}$ for one year to avoid additional progression of hydration during
120 the subsequent tests.

121

122 2.2 Aggregate properties

123 Short-term length change isotherms of the aggregates were determined with a
124 humidity-controlled thermo-mechanical analyzer (BrukerAXS TMA4000SA with HC9700) to
125 study volume changes of the aggregates. Three samples with dimensions of $3\text{ mm} \times 3\text{ mm} \times 6$
126 mm were cut from each aggregate specimen with a diamond saw in three orthogonal directions
127 taking into account their anisotropy. The original aggregates were the largest among the
128 aggregate batches. Length changes were measured with specimens placed under controlled RH
129 levels of 80%, 60%, 40%, and 20% at 20°C for four hours. A linear variable differential
130 transformer with a precision of $0.5\ \mu\text{m}$, a resolution of $0.0025\ \mu\text{m}$, and a contact load of 0.098
131 N was used to measure changes in the lengths of the samples.

132 One sample was analyzed for each direction and each aggregate type; therefore, our
133 experimental results cannot be considered as being representative values. Despite this, our
134 results confirm a difference in shrinkage properties of LS and SS.

135 The Young's modulus and Poisson's ratio of the aggregates are also considered an important

136 factor for preventing large shrinkage of the cement paste [11]. Therefore, these values were
137 calculated by measuring the ultrasonic velocity of the aggregates. Ultrasonic pulse velocities
138 of the P-wave (longitudinal elastic wave) and the S-wave (transverse elastic wave) of
139 water-saturated aggregate samples were measured using an ultrasonic probe (V103-RM and
140 V153-RM, Panametrics-NDT), and a pulsar-receiver (5077PR, Parametric-NDT). The
141 voltage of the pulse oscillator was -400 V, the frequency was 1.0 MHz, and the pulse
142 repetition frequency was 100 Hz for the transmission method. The width of the samples was
143 measured as being 10 mm with a digital micrometer caliper with an accuracy of 0.020 mm.
144 Reference curves were obtained by direct contact, and the period of the pulse peak in the
145 reference curve was subtracted from the period of the pulse peak in the sample record to
146 determine the propagation time. The pulse velocities of the P-wave (V_p) and S-wave (V_s)
147 were calculated from the sample width and propagation time. Using the saturated aggregate
148 density (ρ), Poisson's ratio and Young's modulus were determined by using V_p and V_s
149 according to the following equations:

150

$$151 \quad \nu = \frac{1 - 2(V_s / V_p)^2}{2 - 2(V_s / V_p)^2} \quad (1)$$

152

$$153 \quad E = V_p^2 \rho \frac{(1 + \nu)(1 - 2\nu)}{1 - \nu} \quad (2)$$

154 Results of the three samples and three measurement times for each sample were averaged.
155 One aspect of the background of this experiment is addressed here. The dynamic
156 measurement result for the modulus of elasticity does not always correspond to the static
157 loading result, and the value obtained by the dynamic method is generally larger than that
158 obtained by the static loading test [51]. This is generally explained by the presence of fine
159 cracks in the rock and this tendency is likely to be found in cases where the specimen size is

160 large. The authors agree that the static loading test is more suitable than the dynamic method,
161 however due to size limitations the authors selected the dynamic method to consider the
162 properties of the aggregate.

163

164 2.3 Unrestrained shrinkage experiment

165 Strain distribution in a section of a concrete specimen during the drying process was evaluated
166 by DICM, accompanied by linear deformation and mass change measurements. In the present
167 study, we focused on the section perpendicular to the drying surface, and thus a water vapor
168 impermeable coating to the sides of the specimens was applied.

169 Concrete samples were placed in a steel mold with a diameter of 100 mm and a height of 200
170 mm. After a one-year curing period, disks with a diameter of 100 mm and a height of 9 mm
171 were cut out with a diamond saw. Two circular surfaces were coated with a two-part epoxy
172 resin (Kikusui primer EPW, Kikusui Co.) to form a white vapor impermeable film as shown in
173 Fig. 3. Preliminary testing, which consisted of periodic mass change measurements of a
174 concrete sample fully coated by this resin, confirmed that the coating was sufficiently
175 impermeable to water vapor and elastic enough to not affect the volume change of concrete.
176 The details are introduced in Appendix. Black acrylic ink was sprayed on the test surfaces to
177 create a random pattern consisting of dots with a diameter ranging from 10 μm to 100 μm for
178 the digital image correlation analysis. Specimens were dried under conditions of $20^\circ\text{C} \pm 2^\circ\text{C}$
179 and $60\% \pm 5\%$ RH. Changes in the length of the specimens were measured with the micrometer
180 MHN3-25MB (Mitsutoyo Co.) with a resolution of 0.001 mm and a precision of ± 0.003 mm,
181 and the corrected length of specimens was obtained by calculating the difference in length with
182 respect to a reference stainless steel bar. Specimens were subjected to the first length
183 measurement before drying followed by subsequent measurements every few days. Measured

184 values of three diameters were averaged and recorded as the drying shrinkage strain. All
185 measurements were executed in a room at a temperature of $20^{\circ}\text{C} \pm 2^{\circ}\text{C}$.

186 Changes in mass were measured with a precision balance with an accuracy of 0.04 g at the
187 same time as the length measurement, and the rate of change was determined with respect to
188 the initial mass. Each condition had three samples and averaged values are used for the
189 discussion except for the DICM image results shown in Fig.9 and 10. In Fig.9 and 10, the
190 sample most representative of typical results from the three samples is shown.

191

192 2.4 Details of DICM

193 Digital image correlation (DIC) measurements were performed with a CCD camera Atik
194 383L+ (Artemis CCD Ltd., 3326x2504 pixel) and an Ai AF Nikor 35mm f/2D lens (Nikon Co.)
195 as shown in Fig. 4. A reference image was captured before drying. In this setup, each pixel had
196 a length of 0.043 mm [44, 50].

197 A commercial program (Vic-2D, Correlated Solutions, Inc.) was used for the DIC analysis. An
198 algorithm for maximizing a normalized cross-correlation criterion (NCC) between the
199 deformed subset and the reference subset was implemented in Vic-2D. A subset of 25 pixels x
200 25 pixels, a step of 5 (5-pixel-spacing between centers of subset), and a decay filter (90%
201 center-weighted Gaussian filter) with a size of 15 were applied for conducting DIC to
202 determine local displacement and strain distributions. A cross-section containing entrained and
203 entrapped air bubbles on the specimen surface was omitted in the image analysis due to the
204 occurrence of defocusing and shadow dropping in air bubbles causing inaccuracies in DIC
205 results. This can sometimes cause the abortion of the DIC calculation itself. Even small cracks
206 on the concrete surface can produce a large expansive strain in the DIC algorithm. Parameters
207 were set based on data from our preliminary study [44]. We have also demonstrated that a

208 positive maximum principle strain distribution is well reflected by a micro-crack distribution
209 as confirmed by a fluorescent epoxy impregnation method [44].

210 Fine cracks have been detected and observed with a scanning electron microscope (SEM) and
211 fluorescent epoxy impregnation techniques; however, these methods are not suited for
212 observing changes in crack development. In this study, the use of DICM should allow the
213 measurement of the development of strain distribution through discrete data and permit an
214 informed discussion of the behavior of fine cracks.

215

216 2.5 Restricted shrinkage experiment

217 Specimens with dimensions of 100 mm × 100 mm × 400 mm were subjected to underwater
218 curing with a saturated calcium hydroxide solution for a year and then cut out to have
219 dimensions of 185 mm × 100 mm × 9 mm. Specimens were then fixed in a steel frame with
220 two sections of acrylic adhesive as shown in Fig. 5 and subjected to a restricted shrinkage
221 measurement. Similar to the unrestrained specimens, two cutout surfaces were coated with a
222 vapor impermeable film, which permitted water to escape only through the sides of the
223 specimens. Specimens were then subjected to drying at a temperature of $20^{\circ}\text{C} \pm 2^{\circ}\text{C}$ and a
224 RH of $60\% \pm 5\%$.

225 The target surfaces of DIC measurements were prepared by spraying black acrylic ink on the
226 white, impermeable epoxy coating to produce a random pattern. DIC measurements were
227 recorded with the camera system by following the protocol in the previous section 2.3 and 2.4.
228 In the measurement setup shown in Fig. 6, each pixel had a length of 0.079 mm. For each
229 concrete mixture, one specimen was examined. Obtained data were evaluated by comparing
230 against values in the literature and by numerical analysis.

231

232 3. Experimental results and discussion

233 3.1 Unrestrained shrinkage experiments

234 Results of the length change measurement experiments on the disc specimens are shown in
235 Fig. 7. The concrete total strain of SS was approximately 120 μ larger than that of LS after
236 91-day drying. This shrinkage strain difference was more than the theoretical calculated
237 strain difference of 80 μ , which was estimated by multiplication of 200 μ , the difference in the
238 aggregate shrinkage strain (taken from Fig. 2), by 0.4, the volume fraction of the coarse
239 aggregate. This result suggests that the difference in shrinkage behavior of concrete can be
240 mainly attributed to the drying shrinkage strain of the aggregate and that the average
241 aggregate shrinkage difference might be more than 200 μ , while there remains a possibility
242 that the properties of the interfacial transition zone (ITZ) influence the drying shrinkage of
243 concrete [52, 53].

244 Changes in mass shown in Fig. 8 were larger for SS than for LS. This can be attributed to
245 excess water released from the sandstone aggregate, as its water absorption is twice as large
246 as that of the limestone. The results of DIC measurement for LS and SS are shown in Figs. 9
247 and 10, respectively. Based on the literature showing that expansive strains in the maximum
248 principal strain distribution correspond to fine cracks, subsets showing strains larger than 500
249 μ were taken as the areas containing fine cracks [44]. Minimum principal strains obtained
250 with DIC are shown in Fig. 9, where the LS aggregate did not show shrinkage prior to drying.
251 With drying age, changes in color from yellow to purple were observed at mortar parts,
252 suggesting that the shrinking zone developed from the perimeter towards the inside of the
253 specimen. This tendency was also confirmed for SS as shown in Fig. 10. Similar to those
254 observed in the minimum principle strain distribution, some areas showed maximum
255 principal strain distributions changing from yellow to red (i.e., expansive strains developed
256 from the perimeter towards the inside of the specimen, particularly around aggregates). These
257 phenomena likely reflect crack initiation and propagation due to drying and resultant

258 shrinkage strain distribution.

259 Development of numerous fine cracks (i.e., expansive strains) could be confirmed at the
260 center of the LS specimens even at an age of 14 days, while expansive strains in SS were not
261 significant even at an age of 44 days. The differential cracking tendency is a fertile area for
262 further exploration. To discuss this difference in cracking progression, the minimum strain
263 distributions over the test surface as a function of material age are summarized and shown in
264 Fig. 11. The minimum principal strains at 2.5 mm, 11.5 mm, 24.5 mm, 37.5 mm, and 47.5 mm
265 from the original point, averaging over the ± 6.5 mm of each point on 10 radial lines with
266 arbitrarily selected angles, were averaged. The detailed calculation procedure is represented
267 in Fig. 11(c). A gradient of shrinkage strain can be found in a region 35 mm from the drying
268 surface of the LS specimen even at the age of one day. The behavior of the region remained
269 unchanged after 14 days. Taking into account the minimum principal strain distribution
270 shown in Fig. 11(a) and (b), it can be hypothesized that the excessive shrinkage takes place
271 due to accelerated drying caused by fine crack propagation from the surface and around the
272 coarse aggregates, and the aforementioned development of fine cracks originates due to an
273 uneven shrinkage behavior between limestone and mortar. The synergistic impact of crack
274 development and acceleration of drying inside of specimen is represented in Fig. 11.

275 On the other hand, the gradient of shrinkage strain remained within 20 mm from the drying
276 surface in the SS specimens. Taking into account the maximum principal strain distributions
277 shown in Fig. 10, the shrinkage strain gradient can be attributed to an inhibition of drying due
278 to the large shrinkage of aggregate resulting in less uneven changes in the volumes of
279 aggregate and mortar. In addition to the intrinsic shrinkage properties of GS and mortar, the
280 water supply from the aggregate to mortar might help maintain the moisture content of the
281 mortar, resulting in the inhibition of uneven shrinkage between the aggregate and mortar.
282 This mechanism seems likely since the mass change of SS was larger than that of LS as

283 shown in Fig. 8. It has also been proven that water-saturated, porous aggregates can
284 compensate for the drying of surrounding mortar [54, 55].

285 The present study has not experimentally confirmed that the aggregate-mortar ITZ influences
286 the fine crack generation around the aggregate. However, the literature suggests that the
287 hydrophilicity of the aggregate surface may affect crack propagation [56]. Differences in
288 hydrophilicity cause different characteristics of the ITZ, such as the thickness of voids on the
289 surface of aggregates, bond strength, and stiffness of the nominal mortar-aggregate interface,
290 to arise through the influence of the cement particle packing process on the surface aggregate
291 in fresh concrete. However, the result of a numerical analysis of unrestrained shrinkage under
292 ideal conditions showed that the bond strength between aggregate and mortar exerts no
293 significant effect [43]. On the other hand, it has been observed that the surface of the
294 limestone surface is denser than that of sandstone [57], and a larger bond strength with the
295 limestone aggregate surface would be expected [56]. Therefore, the shear stress on the
296 limestone surface is stronger than that on the sandstone surface. In addition to this, cross
297 cracks between aggregates are more probable in the case of limestone aggregates due to
298 stronger bond strengths, hence facilitating easy crack propagation.

299

300 3.2 Restricted shrinkage experiment

301 The minimum and maximum principal strain distributions in the LS and SS specimens are
302 shown in Figs. 12 and 13, respectively. Regions with the maximum principal strain larger
303 than 500 μ were assumed to be origins for possible generation of fine cracks and are shown in
304 orange or yellow in the maximum strain distribution images. Sample recordings during the
305 progression of drying through permeable surfaces are shown from the top to the bottom of the
306 figures. The migration of shrinkage from the periphery towards the inside can be confirmed
307 from the minimum principal strain distributions. The maximum strain distribution images of

308 LS confirm that numerous fine cracks were generated at the beginning of the drying process
309 and that they gradually progressed towards the inside of the specimen. The fine cracks did not
310 show any interlinking with each other, at least at the observed surfaces, and were evenly
311 distributed when the limestone aggregate was used. This can be attributed to the large
312 difference in shrinkage and (likely) large bond strength between the mortar and limestone
313 aggregate.

314 On the other hand, a few fine cracks were present at the surface of the SS specimens that
315 proceeded to combine into a single large through-crack during drying. Differences in
316 shrinkage between the sandstone aggregate and mortar could potentially be controlled by two
317 factors: the intrinsic shrinking properties of the aggregate and the inhibition of the shrinkage
318 of mortar with water released from the aggregate. Less uneven shrinkage behavior between
319 the aggregate and mortar decreases the possibility of crack generation around aggregates. A
320 smaller bond strength between the aggregate surface and mortar also decreases the possibility
321 of cross cracking between aggregates. Assuming these effects, cracks generated at the surface
322 of concrete would accelerate the drying process along the surface of cracks, and consequently
323 promote shrinkage near the cracks. Therefore, cracks are not distributed, and instead a single
324 crack grows towards the inside of the specimen. As a result, shrinkage cracks are integrated
325 and localized, allowing water release for further shrinkage and localization of the crack, while
326 progression of the crack opening may release stress around it and contribute towards closing
327 other cracks and suppress the acceleration of drying around them.

328 This observed behavior of cracking in concrete under restraint conditions is consistent with
329 the results from our previous experimental studies [41, 42]. Therefore, even though the
330 number of specimens is limited in the current study, these present experimental results
331 accurately reflect the typical cracking behavior in concrete as affected by aggregate
332 properties.

333

334 4. Numerical study

335 4.1 RBSN

336 The Rigid-Body-Spring Networks (RBSN) model developed by Kawai [47] has been applied
337 extensively for structural analysis. RBSN deals with crack propagation of concrete directly
338 [48] since it represents a continuum material as an assembly of rigid particle elements
339 interconnected by zero-size springs along their boundaries [47]. Being nonlinear, these
340 zero-size springs can simulate the cracking behavior of a continuum material. In the present
341 modeling, each interface between two rigid particles was divided into several triangles
342 sharing the barycenter of the interfacial plane, with each triangle having three individual
343 springs, one for a normal force and two for orthogonal tangential forces. In existing studies
344 (for e.g., [49]), the interfacial plane has a rotation spring for bearing momentum, while in the
345 present study, several divided triangles with springs for normal forces bearing the momentum
346 acting on the interfacial plane as shown in Fig. 14 were used instead. At the same time, the
347 nonlinearity of normal and tangential springs can take into account the nonlinearity of the
348 rotation behavior on the interfacial plane.

349 The nonlinearity and discrete behavior of the continuum material is emulated by cracks
350 developing at the interfaces of the rigid particles. For this reason, crack patterns and the
351 resultant nonlinear behavior of the target model are significantly affected when a mesh design
352 is employed. To solve this problem, random geometry using Voronoi diagrams was applied
353 [48].

354 Concrete sections under restraint conditions, similar to the results shown in Section 3.2, were
355 subjected to the numerical calculation. To evaluate the impact of aggregate properties in
356 isolation, three different phases, namely the mortar matrix, aggregates, and mortar-aggregate
357 interfaces, were modeled.

358 For the mortar matrix, the tensile behavior of mortar was modeled using linear elasticity to

359 tensile strength, followed by a bilinear softening branch of a 1/4 model [58], as shown in Fig.
 360 15(a). The parameters for mortar behavior in the tension field are the tensile strength f_t , the
 361 tensile fracture energy G_{ft} , and the distance between the Voronoi generators (centroid of rigid
 362 particle) h . The mortar behavior in the compression field is shown in Fig. 15(b), whose S-type
 363 curve is derived from the relationship between stress and volume under hydrostatic pressure
 364 conditions [59]. Tangential springs represent the shear transfer mechanisms of cracked and
 365 uncracked mortar matrices (Fig. 16(a)). The softening process was modeled by the following
 366 equations [59]:

$$367 \quad \tau = \begin{cases} G\gamma & (\gamma < \gamma_f) \\ \max(\tau_f + K(\gamma - \gamma_f), 0.1\tau_f) & (\gamma_f < \gamma) \end{cases} \quad (3)$$

368 where G : shear stiffness (N/mm²), τ_f : shear strength (N/mm²), γ_f : strain at the maximum stress
 369 in shear strain and shear-stress relationship, and K : shear softening coefficient. A linear
 370 relationship between shear strain and shear stress was first assumed until the stress reached
 371 the peak. Following the peak, the softening process was determined by the strain and stress
 372 normal to the plane on which the shear force was acting, while the minimum value was
 373 assumed to be $0.1\tau_f$.

374 The shear strength was defined by the Mohr-Coulomb type criterion (Fig. 16(b)) and is
 375 represented by the following equations [59]:

$$376 \quad \tau_f = \begin{cases} c - \sigma \tan \phi & (\sigma > -\sigma_b) \\ c + \sigma_b \tan \phi & (\sigma < -\sigma_b) \end{cases} \quad (4)$$

377 where c : cohesion parameter (N/mm²), ϕ : angle of internal friction (degree), and σ_b :
 378 maximum shear strength of a normal spring (N/mm²).

379 The softening process of shear springs is a function of normal stress as shown in Fig. 16 (c):

$$380 \quad K = \beta G \quad (5)$$

$$381 \quad \beta = \min(\beta_0 + \chi(\sigma / \sigma_b), \beta_{\max}) \quad (6)$$

382 where $\beta_0, \beta_{\max}, \chi$: parameters for shear softening [59].

383 Shear transfer is reduced when the strain in the normal direction to the plane that shear stress

384 acts upon is in the post-peak region. This process is represented by a coefficient β_{cr} [60] as

385 shown in Eqs. (7) - (9):

$$386 \quad \tau = \begin{cases} \beta_{cr} G \gamma & (\gamma < \gamma_{ft}) \\ \beta_{cr} \max(\tau_f + K(\gamma - \gamma_{ft}), 0.1\tau_f) & (\gamma_{ft} < \gamma) \end{cases} \quad (7)$$

$$387 \quad \beta_{cr} = \frac{\varepsilon_t}{\varepsilon} \exp\left\{\frac{\kappa}{\varepsilon_{tu}}(\varepsilon - \varepsilon_t)\right\} \quad (8)$$

$$388 \quad \tau_{ft} = c - f_t \tan \phi \quad (9)$$

389 where $\gamma_{ft} = \tau_{ft}/G$, ε_t : strain at the peak of normal stress, ε_{tu} : ultimate normal strain when stress

390 attains zero, ε : normal strain, and κ : reduction factor for shear transfer due to cracking.

391

392 Linear elasticity was assumed for the aggregate. In general, the strength of the aggregate is

393 larger than that of mortar, and therefore the strength of the aggregate was not taken into

394 account. In the present study, the amount of drying shrinkage of the aggregate was considered

395 as a parameter. We assumed two different magnitudes of shrinkage, specifically 0 μ and -400

396 μ corresponding to the limestone and sandstone coarse aggregates, respectively. These

397 shrinkage values were designed based on former research of sand stone shrinkage that found

398 that 400 μ was almost the maximum shrinkage at 60% RH in the available sandstones [61].

399 The Young's modulus of aggregates may also affect the cracking behavior through a

400 restraining role for mortar shrinkage. The average Young's modulus of sandstone available in

401 Japan was found to be approximately 65GPa by ultra-sonic pulse velocity measurement [62]

402 and double of this value was used for comparison.

403

404 The interface between the aggregate and mortar (i.e., the ITZ [63]) was modeled explicitly,

405 although quantitative data relating to the ITZ has been scarcely reported. The ITZ is generally
406 considered to be produced by the “wall effect” of the cement particle packing process on the
407 surface of the aggregate [64]. Our ITZ is more porous than normal mortar and different
408 physical properties were expected as a result.

409 In a previous RBSN study dealing with concrete as a two-phase material, the compressive
410 failure of cylindrical concrete specimens was accurately reproduced by taking the average of
411 the physical properties of mortar and the aggregate as the value for the ITZ [60]. However, the
412 tension field properties have not been studied comprehensively, although the tension in the
413 porous ITZ zone must have a large impact on the cracking behavior of concrete.

414 Calculation parameters for tensile strength, Young’s modulus, and fracture energy were
415 adopted based on a literature survey of porosity distribution findings [64-67],
416 nano-indentation results, an SEM analysis [64, 68], and physical properties testing [56, 63,
417 69-71]. As the porosity in the ITZ is more than three times that of normal mortar located far
418 from the surface of the aggregate [64, 68], the Young’s modulus and the tensile strength of the
419 ITZ must be half of the normal mortar under the assumption of a linear or exponential
420 relationship between porosity and physical properties. Therefore, in the present simulation,
421 0.50 and 0.75 times the Young’s modulus and tensile strength of normal mortar were
422 considered for the calculations. With regard to fracture properties, Alexander et al. [72] have
423 reported that the fracture energy of the ITZ is possibly less than 10% of that of bulk cement
424 paste or mortar of dolomite aggregates with ordinary Portland cement and silica fume.
425 Therefore, 0.1, 0.2, and 0.4 times the fracture energy of normal mortar were considered as the
426 ITZ parameters.

427 Adopting these parameters, the different stress-strain relations in the compression and tension
428 fields were applied to the modeling of the ITZ. The schematic is presented in Fig. 17.

429 Former research has found that when a limestone aggregate is used, the texture of cement

430 hydrates in the ITZ is densified [57] and the fracture energy and bond strength of the ITZ are
 431 strengthened [56, 57]. Note that aggregate particle size and surface roughness remain
 432 important factors for ITZ properties [63, 71, 73, 74]. Therefore, these trends of ITZ properties
 433 in the case of concrete containing limestone aggregates should be considered during the
 434 interpretation of a numerical analysis.

435

436 4.2 Truss networks model for mass transfer

437 Water diffusion in the mortar and aggregate was modeled using a random lattice, whose mesh
 438 was defined by a Voronoi diagram, originally developed by Bolander and Berton [75]. Lineal
 439 conduit elements connect the Voronoi generators and special nodes set on boundary surfaces,
 440 which are named “Surface truss nodes”, are the centroid of surfaces of Voronoi mesh facing
 441 the boundary. The schematic of the lattice model is shown in Fig. 18 and both the Voronoi
 442 generators and surface truss nodes are shown.

443 The governing equation of potential flow of Eq. (10) was modeled assuming potential flow in
 444 the linear conduit as described in Eq. (11):

$$445 \frac{\partial w}{\partial t} - \frac{\partial m}{\partial t} \operatorname{div}(K(w) \operatorname{grad} m) = \frac{\partial w_{hyd}}{\partial t} \quad (10)$$

446

$$447 \frac{A_e K(w)}{L} \frac{\partial \mu_1}{\partial t} - \frac{\partial m_1}{\partial t} + \frac{\partial w}{\partial t} \frac{A_e L}{6} \frac{\partial \mu_1}{\partial t} + \frac{A_b K(w)}{d_{env}} \frac{\partial \mu_1}{\partial t} - \frac{\partial m_{env}}{\partial t} = 0 \quad (11)$$

448 where w : volumetric water content (g/mm^3); w_{hyd} : water consumption by cement hydration
 449 (g/mm^3); $K(w)$: water transfer coefficient ($\text{mm}^2/\text{s} \cdot \text{g}/\text{mm}^3 \cdot \text{g}/\text{J}$); μ_1, μ_2 : chemical potential of
 450 water in conduit nodes 1 and 2, respectively (J/g); μ_{env} : chemical potential of water in the
 451 environment (J/g); d_{env} : nominal distance for the boundary condition of water transfer from
 452 the matrix to the environment (mm); t : time (s); and ω : volumetric conversion factor, (2.0 in
 453 case of 2-dimensional flow), A_e : the area of the Voronoi facet between the contiguous nodes i

454 and j, L : length of conduit from node 1 to 2. The third term in left hand formula represents the
 455 flow on the boundary surface. It is assumed that a hypothetical element with conduit length of
 456 d_{env} was set on the boundary surface and this will be shown in eq.(15) . In the present method,
 457 the total volume of the conduits was set as the total volume of the target Voronoi elements
 458 [49]. For the time development, the entire matrix was assembled based on eq. (11) and the
 459 Crank–Nicolson scheme was applied with equidistant time steps.

460 The water transfer model was based on the research of Maruyama et al. [76]. The global
 461 chemical potential of water was considered for the flow potential, and the water transfer
 462 coefficient of hardened cement paste $K(w)$ was derived from experimental data. The results
 463 were reproduced by the following equations:

$$464 \quad K(w) = \frac{1}{(5.0 - 9.1R + 4.15R^2)} K_{60} \quad (12)$$

$$465 \quad K_{60} = 1.47 \cdot 10^{-13} \exp(4.41t_w) \quad (13)$$

$$466 \quad t_w = w_g / \rho_w / S \quad (14)$$

467 where K_{60} : reference water transfer coefficient where the relative water content was 0.6
 468 ($\text{mm}^2/\text{s} \cdot \text{g}/\text{mm}^3 \cdot \text{g}/\text{J}$); R : relative water content (-); t_w : statistical thickness of adsorption (nm);
 469 w_g : mass water content where the reference state is oven-dry conditions at 105 °C (g/mm^3);
 470 ρ_w : density of liquid water in the mortar or aggregate ($0.001 \text{ g}/\text{mm}^3$); and S : water vapor BET
 471 surface area (mm^2/mm^3). In the present study, the water transfer coefficient of hardened
 472 cement paste was assumed to be the same as that of mortar.

473 The following equation was used for this boundary condition:

$$474 \quad J_{w,bnd} = A_b K(w) \frac{\mu - \mu_{env}}{d_{bnd}} \quad (15)$$

475 where $J_{w,bnd}$: flux at the boundary (g/s); A_b : area of the finite area on the boundary (mm^2);
 476 μ_{env} : global chemical potential of water vapor of the environment; and d_{bnd} : imaginary
 477 distance from the boundary to the environment (3 mm).

478 For the properties of the aggregate and the ITZ, the water capacity dw/du was based on
479 preliminary experimental data [62], and the water transfer coefficient of the aggregate was
480 assumed to be 10 times that of mortar since the aggregate reaches equilibrium faster than
481 hardened cement paste according to previously measured sorption isotherms [50]. Cracked
482 ITZ, which is expected to have a larger water transfer coefficient, was not considered in the
483 present calculation. This assumption may produce conservative results of cracking behavior
484 in concrete in terms of how it is affected by aggregate properties.

485 In the calculation, the moisture related properties of mortar are estimated from the water to
486 cement ratio of the mixture proportion. Uniformity of the mortar matrix is assumed because a
487 viscosity improver is used in the reference concrete. There is an additional possibility that the
488 water in the aggregate can move during the young age of the sample due to osmotic pressure
489 caused by the ion concentration of pore solution [55] but this phenomenon is considered
490 negligible.

491

492 4.3 Analysis outline

493 In the present study, cracks that might be affected by a presence of aggregate are discussed by
494 RBSM analysis results. Due to the limitations of meshing geometry and the calculation
495 process of RBSM, the target cracks are yielded according to the representative mesh size,
496 which is about 5 mm in the present study as shown in Fig. 19. In other words, the cracks
497 within 5 mm intervals can not be shown directly in the present calculation, and the physical
498 role of these cracks are numerically represented by a reduction of spring stiffness, which is a
499 function of its strain. For this reason, complete reproduction of concrete behavior, which
500 shows true multi-scale cracking and resultant change in macroscopic physical properties, and
501 quantitative evaluation of the reproducibility of the present calculation are impossible
502 because applicable quantitative indices can not be obtained. However, qualitative evaluation

503 gives insight into understanding the role of aggregate in concrete with regard to the crack
504 propagation process. Therefore, the authors attempt to obtain the key parameters of the
505 aggregate in cracking behavior through parametric studies. Although these parametric
506 studies are discussed by relative comparisons, the parameters used in the calculations are set
507 to be as realistic as possible.

508

509 Specimens with dimensions of 200 mm × 100 mm × 9 mm under restraint conditions
510 discussed in Section 3.2 were the target of the present numerical study. For the calculation,
511 the quasi-two dimensional mesh shown in Fig. 19(a) was used. Voronoi meshing with a
512 representative diameter of 5 mm was applied in the X-Y plane, while the same section was
513 held in each element in the Z-direction to better understand the crack propagation process in
514 the specimen.

515 The upper and bottom edges of the model were considered as the boundaries of moisture
516 transport as shown in Fig. 19(b). The environmental conditions were considered to be 20°C
517 and 60% RH. The boundary conditions for force equilibrium and the restraint body of
518 stainless steel bars were modeled by a large spring whose ends were connected to a rigid plate
519 adjoining the edge of the concrete specimen. The stiffness of the spring was calculated from
520 the Young's modulus (205 GPa) and sections (32 mm² × 16 mm²) of the members.

521 Calculations were performed until 91 days after drying. The shrinkage of the mortar or
522 aggregate was modeled as a function of the relative water content ($R = w/w_0$) as shown in the
523 following equation:

$$524 \quad \Delta\varepsilon_{sh} = \alpha_{sh} \cdot \Delta R$$

525 where α_{sh} : coefficient of conversion from relative water content to shrinkage strain and R :
526 relative water content (i.e., the ratio of water content w to the maximum water content w_0).

527 The shrinkage of the mortar or aggregate was considered as an isotopic equivalent nodal force

528 in the calculations.

529 In the present study, the creep of hardened cement paste was not considered since our
530 preliminary experiments showed that the tensile creep coefficient of hardened cement paste
531 was only 0.1 [77]. Further, given the small tensile stress in the present study, the creep strain
532 in the hardened cement paste or mortar should not have a large impact on the calculation
533 results. In our analytical hypothesis, tensile creep of concrete can be explained by the fine
534 cracks due to stress and drying shrinkage and the resultant reduction of Young's modulus of
535 concrete [50].

536 Material properties and parameters used in the calculations are summarized in Tables 5 - 7. In
537 addition, a summary of parameters for numerical analysis and a companion group showing
538 the objectives and notations of the parameter sets are presented in Table 8, where $\epsilon_{sh,XX}$ is
539 shrinkage of XX microns; $E_{a,XX}$ is Young's modulus of the aggregate of XX GPa, $E_{ITZ,XX}$ is
540 Young's modulus of the ITZ of XX times the Young's modulus of mortar, $f_{t,ITZ,XX}$ is tensile
541 strength of the ITZ of XX times the tensile strength of mortar, and $G_{f,ITZ,XX}$ is fracture energy of
542 the ITZ of XX times the fracture energy of mortar.

543

544

545 4.4 Numerical analysis results and discussion

546 4.4.1 Moisture transfer

547 Fig. 20 shows the results of the drying process. As the analysis takes into account differences
548 in the water capacity and the water transfer coefficient between the aggregate and mortar,
549 oscillation in drying depth was observed. After 91 days, almost exclusively within 1 or 2 mm
550 from the surface, the mortar attained equilibrium with the surrounding environment. On the
551 other hand, the center of the specimen still indicated more than 74% of RH.

552

553 4.4.2 Cracking behavior

554 (1) Reproduction of the experimental trends

555 Fig. 21 represents the time-dependent cracking behavior under drying of Sh0_E135_0.4Gft
556 and Sh400_E65_0.1Gft. No cracking was observed until the first day of drying. At three days
557 after drying, several fine cracks were observed on the top and bottom edges of the specimens.
558 In the case of Sh400_E65_0.1Gft, a surface crack propagated along the aggregate surfaces,
559 and a relatively large crack was observed on the upper-left part while many cracks stayed near
560 aggregates in the case of Sh0_E135_0.4Gft. These observations can be explained by the large
561 shrinkage of the aggregate and the small fracture energy of the ITZ. At seven days after
562 drying, Sh0_E135_0.4Gft exhibited a crack distribution with one growing from the top left,
563 and the other growing from the bottom right. On the other hand, Sh400_E65_0.1Gft showed a
564 through-crack. This can also be explained by the large shrinkage of the aggregate and the
565 small fracture energy of the ITZ. A large aggregate shrinkage enhances crack propagation
566 along the aggregates since they shrink during the drying process and a larger stress becomes
567 localized on the surface of the aggregates. This tensile stress promotes crack propagation
568 along the crack surface. Furthermore, the smaller fracture energy of the ITZ means that crack
569 propagation reduces the fraction of the concrete specimen that can bear the total
570 shrinkage-induced stress produced by a restraining body. Therefore, cracks can easily grow
571 under restraint conditions at their front. Thus, the smaller fracture energy of the ITZ has an
572 impact on the localization of cracking.

573 The cracking pattern in concrete is determined by both fine cracks bridging aggregates and
574 wide cracks propagating and connecting the ITZ zones of aggregates. This is similar to the
575 phenomena observed in Section 3.2. In the crack pattern present after 91 days of drying, crack
576 localization is very intense in the case of Sh400_E65_0.1Gft. The close-up figures (Fig. 22)
577 confirm wider small cracks around the aggregate in the case of Sh0_E135_0.4Gft. These

578 results imply that Sh0_E135_0.4Gft transformed the elastic energy accumulated by
579 restraining of shrinkage into multiple fine cracks around the aggregate, while
580 Sh400_E65_0.1Gft did so by localizing one large through-crack. These modeled tendencies
581 reproduced the experimental results in Section 3.2. In the next section, the contribution of
582 each parameter to crack localization is discussed.

583 (2) Impact of individual parameters

584 Fig. 23 shows cracking behaviors after 91 days of drying, as affected by differences in
585 Young's modulus and aggregate shrinkage. In this figure, the Young's modulus of the
586 aggregate had little effect on the cracking pattern in concrete under restraint conditions. This
587 can be explained by the fact that almost all the mortar was under the tension field in the
588 X-direction, and the restraining role of the aggregate for mortar shrinkage did not have a large
589 impact in contrast to the case of free shrinkage [78].

590 On the contrary, the shrinkage of the aggregate had a large impact on cracking behavior in
591 concrete. In the case where the aggregate showed low shrinkage, small cracks were
592 distributed around the aggregate while the concrete containing an aggregate with large
593 shrinkage exhibited one large crack. Thus, the more similar the aggregate and mortar were in
594 terms of their shrinkage properties, the greater was the localization of cracking in concrete
595 under restraint conditions.

596 Fig. 24 shows the impact of the tensile strength and Young's modulus of the ITZ on the
597 cracking pattern. Large cracks appeared in a different position for the Sh0_0.75E_0.5f_t
598 condition. **The results indicated that if we increase the tensile strength of ITZ with constant**
599 **fracture energy, localization of cracking is confirmed.**

600 In Fig. 25, cracking patterns in concrete as affected by the fracture energy of the ITZ are
601 shown. During the crack development of cracking, initial crack patterns among SH0_0.1Gft,
602 Sh0_0.2Gft, and Sh0_0.4Gft were similar almost the same until they were dried for 1 day,

603 because the mesh geometry isare common. However after 1 day drying, But after that, grown
604 cracks were differentiated. The smaller the fracture energy was, the more localized and wider
605 the cracks were. The difference in cracking patterns was largest between Sh0_0.1Gft and
606 Sh0_0.2Gft.

607 Crack distribution evaluated quantitatively is shown in Fig. 26. It shows the frequency of
608 springs assorted by crack widths in a logarithmic scale. Fig. 26(a) shows the crack
609 distribution for aggregates with different shrinkages, Fig. 26(b) shows that for different
610 strength of ITZ, and Fig. 26(c) shows the crack distribution for different fracture energies of
611 the ITZ. In Fig. 24(a), the concrete with a smaller aggregate shrinkage value shows a high
612 frequency of cracks with widths ranging from 0.001 mm to 0.01 mm while large cracks of the
613 order of 0.1 mm show a low frequency. Thus, smaller aggregate shrinkage apparently
614 distributes energy into small cracks of 0.01 mm ~ 0.001 mm in width by way of compensation
615 for cracks of the order of 0.1 mm in width. The same trend was observed in the cases of
616 Sh0-0.1Gft and Sh0-0.2Gft.

617 These analytical studies confirmed that the localization of cracking becomes increasingly
618 apparent when aggregate shrinkage is larger, strength of ITZ is larger, or the fracture energy
619 of the ITZ is smaller.

620 As discussed in the earlier sections, aggregate type has a large impact on the properties of the
621 ITZ and aggregate shrinkage that governs shrinkage-induced cracking in concrete under
622 restraint conditions. Consequently, it can be concluded that the coarse aggregate of pure
623 limestone, which shows smaller drying shrinkage and may densify the ITZ, can reduce the
624 number of visible cracks in concrete under restraint conditions. This is since it allows fine
625 cracks around the coarse aggregate that absorb the localization of cracking.

626

627

628 5. Conclusion

629 Concrete specimens with aggregates having different properties including drying shrinkage
630 were prepared and a water-impermeable coating was applied to control the drying direction.
631 Changes in the shrinkage strain distribution of the cross-section of the specimen
632 perpendicular to the drying direction were observed with a digital image correlation method
633 (DICM) under restricted and unrestrained conditions.

634 The drying shrinkage of the aggregate was found to play a dominant role in determining the
635 drying shrinkage of concrete. The DICM confirmed that when limestone aggregates with a
636 small drying shrinkage were used under unrestrained conditions, the difference in the drying
637 shrinkage between the aggregate and the mortar caused cracks around the aggregate, forming
638 shrinkage strain gradients from the drying surface to the inside. This tendency was smaller
639 when sandstone aggregates with a larger drying shrinkage were used. This was partly due to
640 the porosity of the sandstone aggregate, which allowed it to release excessive water to
641 suppress drying shrinkage during the early stages of drying.

642 Under restricted conditions, specimens with the limestone aggregate showed discontinuous
643 fine cracks developing both at the surface and into the interiors. Meanwhile, for specimens
644 with the sandstone aggregate, fine cracks were distributed over the surface while a single
645 large crack extended inside with time.

646 With the aid of numerical analysis, parameters that could contribute to cracking behavior in
647 concrete, such as the Young's modulus and aggregate shrinkage, strength, stiffness, and
648 fracture energy of the ITZ, were studied. Aggregate shrinkage and the fracture energy of the
649 ITZ were found to govern shrinkage-induced cracking of concrete under restraint conditions.

650 Based on both the experimental and numerical analysis results, it can be concluded that when
651 the difference in drying shrinkage between the aggregate and mortar is considerable, or the
652 fracture energy of the interfacial transition zone (ITZ) is very large, the distribution of fine

653 cracks contributes to the suppression of macroscopic cracks. However, when the difference is
654 small or the fracture energy is small, the development of a single large crack, promoted by the
655 associated drying progression, becomes significant. This consequently leads to the formation
656 of a localized macroscopic crack. Pure limestone, which shows smaller drying shrinkage and
657 may densify the ITZ in concrete under restraint conditions, allows fine cracks to form around
658 coarse aggregate particles that absorb stress and limit crack localization, and thus control
659 macroscopic cracks.

660

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671

672 Appendix

673 A.1 Coating material performance

674 The impervious coating used in this study is compared with the normal concrete surface by
675 evaporation mass per surface area. Environmental conditions during the experiment were 20
676 °C and 60±5% RH. The coating material itself also showed some mass change under these
677 conditions and the material coated on the metal surface was also measured. The results are

678 summarized in Fig. A-1. Based on this figure, approximately 94% of vapor evaporation from
679 the concrete surface was prevented until 150-day-drying by using the coating material. The
680 slower evaporation rate introduced a smaller water content gradient in the specimen and can
681 mitigate cracking on the concrete surface due to large shrinkage differences derived from a
682 steep water content gradient.

683 For this reason, the impact of water vapor evaporation from the coating is considered as
684 negligible for surface cracks caused by the internal restraint in the present study.

685

686

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