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The effects of carbonation conditions on the physical and microstructural properties of recycled concrete coarse aggregates

Asghar Gholizadeh-Vayghan ^{a,b*}, Annelie Bellinkx ^c, Ruben Snellings ^a, Bram Vandoren ^c, Mieke Quaghebeur ^a

^a Sustainable Materials Management, VITO; 200 Boeretang, 2400 Mol, Belgium

^b Faculty of Civil Engineering, K. N. Toosi University of Technology; 1346 Valiasr St, 15875 Tehran, Iran

^c Faculty of Engineering Technology, Hasselt University, Martelarenlaan 42, 3500 Hasselt, Belgium

* Corresponding author: contact at asghar.gholizadehvayghan@vito.be

Abstract – In this research, the effects of carbonation conditions on the physical, mechanical, microstructural and durability properties of recycled concrete aggregate (RCA) are investigated. The results suggest that regardless of the carbonation conditions, carbonation of RCA under wet moisture condition or moist carbonation atmosphere results in no meaningful drop in water absorption or porosity. However, carbonation of RCA (preconditioned at $\leq 95\%$ relative humidity) in the absence of external source of water vapor results in significant drops in water absorption (up to 27%), porosity (27%), micro-Deval wear factor (40% regression) and freeze–thaw resistance (77% regression). Carbonation of RCA under optimal conditions was found to densify the exterior surface of hydrated cement within the RCA grains through progressive deposition of carbonates on the exterior of RCA grains. Carbonation also transforms the transition zone between the old cement and aggregates and results in formation of a dark rim around alite and belite grains indicating reactivation of the residual unhydrated cement fraction.

Keywords: Recycled concrete aggregate; Carbonation; Water absorption; Porosity; Resistance to wear; Resistance to freezing and thawing

1 Introduction

The construction and demolition (C&D) industries are accountable for the largest portion of anthropogenic residues in the European Union [1]. According to the European Commission, 25 – 30% of the total wastes generated in the EU is from C&D activities [2] with concrete being on top of the

list. In terms of quantity, this accounts for nearly 450 million tons of C&D waste produced each year [3]. This figure is expected to grow rapidly as the existing infrastructure approaches the end of their service lives.

Statistics suggest that considerable improvement has been made in the last few years regarding the recovery of C&D waste in the EU region. For instance, according to a EuroStat report (CEI_WM040) only 17% of such wastes were recovered in Belgium in 2010 while this number increased to 95% by 2016. The source also reports that the average recovery rate of C&D waste reached an 89% record in the same year for the 28 member states [4]. While many member states have taken effective measures to improve the recycling of different C&D wastes, it should be noted that, according to the Waste Framework Directive, the term “recovery” is applied to re-using, recycling or subjecting to material recovery, including through backfilling operations [5]. Such definition appears to have led to an inflation in the recovery rate figures. For instance, according to Mulders (2013) 92% of concrete was recovered in the Netherlands in 2012. However, only 20% of the recovered concrete was indeed reused in the concrete industry while the rest was used as road base materials [6]. The capacity of the road construction sector to absorb concrete rubbles as base and sub-base materials is limited in mature infrastructure systems. It is thus imperative to further investigate how recycled concrete aggregates (RCA’s) are recovered, what challenges there are for re-using RCA in concrete applications, and how these can be mitigated. A short discussion thus follows.

The common practice of recycling concrete into new concrete ingredients involves crushing steps to convert the concrete rubbles into aggregates [7]. Two or more RCA fractions are generated during crushing, a fine fraction below 4 mm, and one or more coarse fractions. The fine RCA is usually not recommended for making new concrete [8,9,10] due to its significant fraction of hydrated cement paste, which results in excessively high porosity and water absorption [11,12], rapid loss of workability over time when used in mortar/concrete, and increase in drying shrinkage [13].

The coarse RCA is also known to have inferior properties compared to fresh aggregates (due to the presence of hydrated cement paste) [14]. Nevertheless, coarse RCA is known to be generally suitable for concrete manufacture if only replaced for a fraction of concrete aggregates [8]. Li et al. [15] reported ~13% drop in the 28-d compressive strength of concrete as a result of full replacement of coarse aggregates with recycled concrete coarse aggregates. Other studies suggest that up to 30% replacement of the concrete aggregates with the coarse RCA does not result in a meaningful drop in strength [16,17,18]. Some new guidelines allow for higher rates of replacement (e.g., [19]). However, they limit the high levels of allowances to low-grade concrete used in less demanding applications or exposure classes.

In order to enable a more substantial high-grade application of RCA in concrete, researchers have proposed different methods to improve the water absorption and surface properties of RCA. Surface

67 coating of RCA with a pozzolanic slurry [20,21,22], poly-vinyl alcohol polymer [23], and water glass
68 [21,24] have been explored in the past with satisfactory results. Shi et al. (2016) [11] reviewed
69 different RCA treatment techniques reported in the literature and concluded that carbonation is an
70 efficient and environmentally friendly method for enhancing the properties of RCA.

71 Indeed, carbonation of RCA is gaining more attention among researchers due to its positive influence
72 on the physical and mechanical properties of the aggregate, ease of application, and environmental
73 implications [25,26,27]. In the research by Li et al. [15] where replacement of aggregates with non-
74 carbonated RCA was found to cause ~13% drop in strength, similar concrete mixtures with
75 carbonated RCA showed no drop in strength compared to the reference mixture. The authors applied
76 carbonation on air-dried RCA under 100% CO₂ concentration at ambient pressure for 7 days. Zhang
77 et al. [28] also reported comparable compressive strength results for the reference concrete mixtures
78 and the mixtures incorporating carbonated RCA (20% CO₂ concentration for 7 days), while that of the
79 non-carbonated RCA was 8 – 15% less than the reference.

80 The calcium-bearing hydration products (portlandite, C–S–H, ettringite and AFm phases) in the
81 hardened cement paste are prone to carbonation. In the case of portlandite, carbonation leads to 11 –
82 12% increase in solid volume [27,29,30]. This can be correlated to a decrease in the porosity of the
83 old cement matrix and an increase in the mechanical strength of the RCA [27,30]. On the other hand,
84 carbonation of C–S–H is a complex process with implications on the porosity which are still subject
85 to debate [31,32,33]. Some researchers are convinced that carbonation of C–S–H leads to 23%
86 increase in volume and contributes to reduction in water absorption [11,27] while others refrain from
87 reiterating such value or the perceived increase in volume as a whole [33]. Nonetheless, the
88 carbonation of ettringite entails as much as 50% volumetric shrinkage as a result of full carbonation
89 per reaction (1). As such, care should be taken when choosing the carbonation conditions to maximize
90 the reduction in RCA porosity.



91 A number of factors during the carbonation process seem to affect the porosity of carbonated RCA.
92 The CO₂ pressure, ambient temperature and relative humidity, carbonation duration, and aggregate
93 moisture condition appear to be the governing carbonation factors. However, most of the existing
94 literature lends itself to the investigation of carbonation under ambient or close to ambient
95 atmospheric pressures. Zhan et al. (2014) [30] reported a 16.7% drop in water absorption of coarse
96 RCA after exposure to accelerated carbonation settings (low relative humidity, 10 kPa CO₂ pressure,
97 and an ambient temperature of 23 °C). Zhang et al. (2015) [29] reported 23 – 28% drop in such
98 parameter after carbonating aggregates at 20 °C, 60% RH and 20% CO₂ concentration (results varied
99 depending on aggregate type). Considerable improvements in the mechanical properties of RCA (e.g.,
100 aggregate crushing value, impact value and Los Angeles abrasion value) as a result of carbonation has

101 also been reported [29,34]. The research by Shi et al. [11] suggests that if CO₂ pressure exceeds 0.5
102 MPa (~5 bars), the strength development of carbonation products will be insignificant. The authors
103 suggested that this is probably because the performance enhancement obtained through carbonation of
104 portlandite is counteracted by the deterioration of other cement hydrates such as C–S–H. A similar
105 conclusion is drawn by Bertos et al. (2004) [32] and pressures exceeding 5 bars were deemed
106 impractical. As such, it seems reasonable to limit the carbonation pressure to 5 bars.

107 Research on the effect of carbonation temperature is particularly scarce as Kaliyavaradhan and Ling
108 (2017) concluded in their review paper [35]. Bertos et al. (2004) [32] argue that while the CO₂ uptake
109 increases with increase in temperature up to 60 °C, due to the exothermic nature of the carbonation
110 reaction, increase in temperature promotes formation of meta-stable forms of C \bar{C} . The more stable
111 forms of C \bar{C} will form at much lower temperatures (0 – 10 °C) [32]. Moreover, carbonation is
112 commonly reported to be the fastest when the relative humidity is 40 – 70% and most research on
113 carbonation of RCA has been conducted at RH in this range [32,33,36], whereas some other
114 researchers have reported good results for carbonation of RCA at very low relative humidity (e.g.,
115 [30]). It appears that the suitable chamber RH is dependent upon aggregate moisture conditions as
116 well, which has been mostly overlooked in the past. As such, finding the optimal carbonation
117 conditions requires further research.

118 The main objective of this research is to investigate the effects of RCA moisture condition and
119 carbonation conditions (relative humidity, CO₂ pressure, temperature, and duration) on the physical,
120 microstructural, mechanical and durability properties of coarse RCA. The main parameters
121 investigated include the density, water absorption, porosity, micro-Deval resistance to wear,
122 resistance to freezing and thawing, depth of carbonation and the microstructure of laboratory-made
123 RCA. The objective of this research is to (1) evaluate the influence of the above variables on the
124 absorption and porosity of RCA, (2) identify the outstanding carbonation conditions, and perform a
125 more in-depth investigation on the microstructure, surface wear resistance and freeze-thaw resistance
126 of RCA produced under such conditions, and (3) interpret the observations to extend the
127 understanding on the effects of carbonation on RCA properties.

129 **2 Methodology**

130 A base concrete mixture is first produced, cast and cured inside sealed plastic buckets for production
131 of RCA. After 28 days, the concrete is first crushed into hand-sized rubbles using a point-press
132 hydraulic jack. The rubbles are dried to constant mass and then crushed using a laboratory jaw
133 crusher. The >3 mm fraction is tested for its apparent particle density and saturated surface-dry
134 density, water absorption, micro-Deval (MDE) and freeze-thaw resistance. Samples of this fraction

are then subjected to different carbonation conditions where the RCA moisture condition, chamber relative humidity, CO₂ pressure, temperature, and carbonation duration are studied in two to four levels in a sequential evolutionary design of experiments. The carbonated RCA specimens are then tested for the same properties and the results of each experiment are used for deciding on the next carbonation conditions. Satisfactory results are obtained after studying 16 different carbonation conditions. A simple closed-form formula for determining aggregate porosity as a function of its apparent and oven-dried density is derived and the optimal combination of variable levels for maximum drop in porosity and water absorption is finally determined. The products of outstanding carbonation conditions are studied for depth of carbonation via the phenolphthalein spraying technique. The RCA microstructure and morphology are also studied using the scanning electron microscopy technique. The MDE and freeze-thaw resistance of the RCA carbonated under optimal settings is then measured and compared to those of fresh aggregates and non-carbonated RCA.

Materials

Neat Portland cement (CEM I 52,5 N) is used as the primary (and only) cementitious material in making the base concrete. 0/3 siliceous river sand, 2/7 and 7/14 crushed limestones are also used as the sources of fine aggregates, small and large coarse aggregates, respectively (the numeric designations pertain to the aggregate nominal sizes (min/max) in mm based on EN 12620). Table 1 shows the mixture proportions of the base concrete. A high effective water-to-cement ratio of 0.57 is chosen to promote high water absorption by RCA and to ensure the applicability of the research findings to extreme situations where extra water is added to concrete to help mixing and compaction.

Table 1. Mixture proportions of the base concrete.

Cement (kg/m ³)	Water (kg/m ³)	0/3 sand (kg/m ³)	2/7 limestone (kg/m ³)	7/14 limestone (kg/m ³)
360	205	535	535	710

The gradation of the 0/3 siliceous sand and 7/14 limestone are used to determine the ideal crushing steps to generate RCA with particle size distribution similar to those of the starting materials. The physical properties of aggregates are also listed in

Table 2. The water absorption of 2/7 and 7/14 aggregates are below 1.0% while that of the 0/3 sand is no more than 0.37%. The surface wear resistance of 7/14 limestone is also tested in accordance with EN 1097-2 (MDE resistance to wear, also called MDE factor), which was found to be $16.0 \pm 0.52\%$ (note that the reported error margins all pertain to 90% confidence interval, i.e., $6.31 \times$ standard deviation throughout the paper).

167

168

Table 2. Physical and mechanical properties of fresh aggregates.

Aggregate (size range)	Apparent particle density (kg/m ³)	Oven-dried density (kg/m ³)	Water absorption (%)	Porosity* (%)	MDE factor (%)
7/14	2715	2669	0.64	1.73	16.0 ± 0.52
2/7	2701	2643	0.82	2.22	NA
0/3	2658	2632	0.37	0.99	NA

* The porosity values are obtained via the closed-form formula derived in the results section.

170

171 Methods

172 Production of RCA

173 Upon completion of mixing, representative samples from the fresh base concrete mixture are taken
 174 and moist cured inside three 15×15×15 cm³ cubic specimens for 28 days and tested for their
 175 compressive strength per EN 12390-3 (28-day compressive strength = 57.6 MPa). The remaining
 176 concrete is stored inside sealed plastic buckets for the same duration of time. They are then crushed
 177 by first crushing to <9 mm rubbles using a hand-driven point-press hydraulic jack, and then to
 178 aggregate size grains using a jaw crusher. It should be noted that crushing concrete to RCA is usually
 179 undertaken in several steps where it is desirable to carry out the primary step using a jaw crusher (due
 180 to its high efficiency). However, jaw crushers tend to produce high amounts of flaky/needle-shaped
 181 grains. As such, the next steps of crushing are usually carried out with a cone crusher or more
 182 preferably an impact crusher to avoid generation of such grains. However, due to lack of access to
 183 such types of crushers, the rubbles are crushed to aggregates only using a laboratory jaw crusher. The
 184 crusher opening is first set at 10 mm opening and all rubbles are passed through. Next, the resulting
 185 grains are crushed once more at 8.5 mm opening. This is found to yield RCA with fairly low
 186 flaky/needle-shaped grains and a particle size distribution similar to those of the starting aggregates.
 187 The obtained RCA is screened for the passing of 3 mm. The experimental program is geared towards
 188 studying the effects of carbonation on the properties of the >3 mm fraction. The grains are
 189 immediately washed after crushing, dried and stored for carbonation procedures.

190 Design of variables

191 Five variables are defined and investigated in this research as follow. RCA Moisture condition
 192 (“Wet”, “Preconditioned”, “Air-dried” and “Oven-dried”), Relative humidity of the carbonation
 193 atmosphere (with or without boosting to saturation; referred to as “Boosted” or “Not boosted”), CO₂
 194 pressure (1 bar or 5 bars), Temperature (20 °C or 60 °C), and carbonation Duration (1 hour, 4 hours or
 195 24 hours). See Table 3 for a detailed description of carbonation variables and how they are realized in
 196 the experiments.

197

Table 3. Detailed description of carbonation variables.

Variable	Levels	Description
Moisture condition (Qualitative)	“Wet”	The RCA is soaked in water for one week, and then drained on sieve 1 mm for 10 minutes before carbonation. Moisture content: 5.6±0.3%
	“Preconditioned”	The RCA is first dried at 80 °C for 1 day and then placed inside a climate chamber set at 95% relative humidity for 48 hours. Moisture content: 1.01%
	“Air-dried”	The RCA is exposed to ambient air with RH=50±5% for 24 hours. Moisture content: 0.73%
	“Oven-dried”	The RCA is dried at 100 °C for 24 hours. Moisture content: ~0.0%
Relative humidity (qualitative)	“Boosted”	500 ml of water is placed inside the chamber 2 hours before carbonation.
	“Not-boosted”	The above step is not taken.
CO ₂ pressure (bar)	1	Rapid increase from ambient CO ₂ pressure (~ 0 bars) to the test level in the carbonation chamber
	5	
Temperature (°C)	20	Tests are run at 20±3 °C ambient temperature.
	60	The RCA and CO ₂ chamber are preheated to 60±3 °C before carbonation.
Duration (h)	1	CO ₂ pressure is maintained at the maximum specified value for the specified duration.
	4	
	24	

Design of experiments (DoE)

A sequential evolutionary set of experiments is designed for studying the effects of the above variables on the properties of RCA after carbonation. The deployed DoE is referred to as ‘sequential evolutionary’ because the levels of variables in each phase of experiments are chosen based on the results of the previous phase(s). Table 4 shows the full set of experiments. The rationale behind carrying out each experiment is provided below. The starting carbonation conditions are chosen as follows (baseline conditions): “Wet” RCA carbonated under “Boosted” RH at 1 bar CO₂ pressure for 1 h at 20 °C. The said conditions are listed under Exp. 2 in Table 4. In the next experiment, the pressure is raised to 5 bars to observe the effect of CO₂ pressure (Exp. 3). Exp.’s 2 and 3 are then repeated at 60 °C to explore the effect of temperature (Exp.’s 4 and 5). The results directed the authors to dial the temperature back to 20 °C and instead investigate the outcome of carbonation under “Not boosted” conditions for longer durations of time at both 1 and 5 bars (Exp.’s 6 to 8). Next, the availability of moisture is further eliminated by switching the RCA moisture condition from “Wet” to “Preconditioned” while the relative humidity is studied under “Boosted” condition (tested at 1 and 5 bars for 1 hour: Exp.’s 9 and 10). After concluding that carbonation does not make notable progress under “Boosted” levels of RH, it is studied under “Not boosted” condition at 1 bars for 1 and 24 hours to see the effects of both elimination of relative humidity and extension of carbonation duration (Exp.’s 11 and 12). After confirming the merits of both of the mentioned changes, the effects of high CO₂ pressure (i.e., 5 bars) is investigated once more under “Not boosted” and “Preconditioned” settings (studied at two levels of carbonation duration: see Exp.’s 13 and 14). The

220 outstanding results obtained during Exp.'s 11 to 14 propelled the authors to further eliminate the
 221 presence of moisture while maintaining the carbonation pressure and time the same (i.e., 5 bars and 4
 222 hours) to see whether further removal of moisture improves the carbonation outcome (Exp.'s 15 and
 223 16).

224
 225 Table 4. The list of experimented carbonation conditions.

Exp. No.	Moisture condition	Relative humidity	Temperature (°C)	Pressure (Bar)	Duration (h)	Label
1	CTRL	–	–	–	–	CTRL
2	Wet	Boosted	20	1	1	NP/W/B/20C/1b/1h
3	Wet	Boosted	20	5	1	NP/W/B/20C/5b/1h
4	Wet	Boosted	60	1	1	NP/W/B/60C/1b/1h
5	Wet	Boosted	60	5	1	NP/W/B/60C/5b/1h
6	Wet	Not boosted	20	1	4	NP/W/NB/20C/1b/4h
7	Wet	Not boosted	20	5	4	NP/W/NB/20C/5b/4h
8	Wet	Not boosted	20	1	24	NP/W/NB/20C/1b/24h
9	Preconditioned	Boosted	20	1	1	NP/PC/B/20C/1b/1h
10	Preconditioned	Boosted	20	5	1	NP/PC/B/20C/5b/1h
11	Preconditioned	Not boosted	20	1	1	NP/PC/NB/20C/1b/1h
12	Preconditioned	Not boosted	20	1	24	NP/PC/NB/20C/1b/24h
13	Preconditioned	Not boosted	20	5	1	NP/PC/NB/20C/5b/1h
14	Preconditioned	Not boosted	20	5	4	NP/PC/NB/20C/5b/4h
15	Air-dried	Not boosted	20	5	4	NP/AD/NB/20C/5b/4h
16	Oven-dried	Not boosted	20	5	4	NP/OD/NB/20C/5b/4h

226 227 **Carbonation procedures**

228 Each recipe of carbonation is initiated by first adjusting the moisture condition of representative RCA
 229 samples as described in Table 3. The samples and the carbonation chamber are then brought to the test
 230 temperature two hours before the test. Next, a nylon tray containing 500 ml of water is placed inside
 231 the chamber in the case of “Boosted” experiments two hours in advance. The samples are then placed
 232 inside the chamber and the carbonation is initiated by quickly injecting CO₂ into the chamber to
 233 achieve the designated CO₂ pressure. The pressure is preserved at that level for the specified duration
 234 and finally released and the chamber is flushed with nitrogen for 2 minutes. Upon completion of
 235 carbonation, the samples are taken out and dried at 80 °C in a ventilated oven for 24 hours to remove
 236 all the water trapped or generated during carbonation.

237 **Test procedures**

238 ***Density and water absorption measurement***

239 The RCA specimens are soaked in water for 24 hours and tested for density and water absorption in
240 duplicates (in most cases) per EN 1097–6. The total porosity of the specimens is also estimated as
241 explained next.

242 ***Porosity estimation***

243 Aggregate porosity plays a significant role in its water absorption, freezing and thawing resistance,
244 and drying shrinkage of concrete. It is thus imperative to find a means to adequately measure/estimate
245 this parameter. The RCA porosity originates from the hydrated cement paste adhering to the grains,
246 which contains gel (0.5 nm – 10 nm) and capillary (10 nm – 10 µm) pores [37]. It is prudent to bear in
247 mind that there is no unified definition for cement porosity, and no existing test method can explore
248 and characterize the entire pore size range of cement. For practical purposes, however, it can be
249 argued that all the pores that can be filled with water in a 24–h period can be effectively considered as
250 porosity. As such, the ratio of such pores volume to the total volume of grains (i.e., the sum of solid
251 matter volume and pore volume) can be considered as the effective porosity. If this definition is valid,
252 the apparent particle density and the oven–dried density values can be used to estimate the effective
253 porosity. A closer look at the definitions of apparent particle density (ρ_a) and oven–dry density (ρ_{od})
254 suggests that the effective porosity of RCA (p) can be easily obtained by dividing the two. The
255 following simple derivations lead to the formula for effective porosity as listed under Eq. 1. Such
256 parameter is calculated for all fresh aggregates as well as the CTRL and carbonated RCA.

$$\rho_{od} = \frac{m_{od}}{V_{ssd}} = \frac{m_{od}}{V_{od} \times (1 + p)} = \frac{\rho_a}{(1 + p)} \rightarrow$$
$$p = \frac{\rho_a}{\rho_{od}} - 1$$

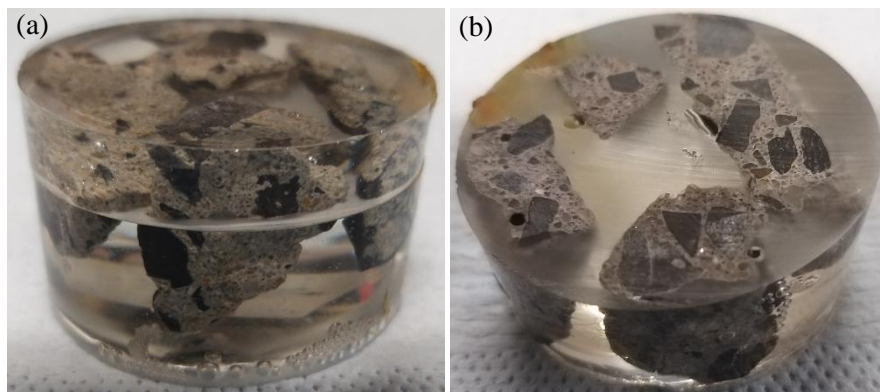
Eq. 1

258 ***Micro–Deval and freeze–thaw resistance measurements***

259 In order to observe the impacts and implications of carbonation on the mechanical and durability
260 properties of RCA, the micro–Deval resistance to wear and the resistance to freezing and thawing are
261 measured on selected aggregates. The tests are run in duplicates on the starting aggregates (i.e., 7/14
262 limestone), the CTRL (i.e., non–carbonated) RCA and the RCA carbonated under optimal settings
263 (judged by the water absorption and porosity results) per EN 1097–1 and EN 1367–1, respectively.

265 **Carbonation depth measurement**

266 The depth of carbonation is qualitatively assessed on RCA with outstanding water absorption/porosity
267 results via the following procedure. Four to six RCA grains are picked at random from the selected
268 carbonation procedures, dried to constant mass and submerged in a low-viscosity epoxy resin inside a
269 4-mm plastic cup. Upon hardening, the epoxy tablet containing the grains is cut using a fine saw such
270 that the grains are cut and exposed approximately from the middle (see Figure 1). The cut surface is
271 rinsed with ethanol, dried and later damped with a 0.5% phenolphthalein solution using an absorbent
272 cloth for detecting the alkalinity of cement hydrates in the RCA. The change in color (if any) is then
273 photographed 15 minutes after exposure.



274
275
276 Figure 1. Sample preparation for carbonation depth determination: (a) epoxy-submerged RCA
277 grains and the cut section; (b) the cut and exposed RCA grains.

278
279 **Microstructural investigations**

280 Three grain are randomly selected from each RCA with outstanding water absorption/porosity results
281 are dry-cut from the middle using a fine saw. The grains are then placed inside a 3-mm plastic cup
282 with the cut surfaces facing downwards and impregnated with a low-viscosity epoxy resin under
283 vacuum. Upon curing, the tablets are gradually polished down to 1 μm diamond powder grit size. The
284 samples are Pt/Pd-coated prior to microscopic analysis. An FEI Quanta 200 scanning electron
285 microscope equipped with a Bruker XFlash 4030 EDS detector is used to explore the morphology,
286 composition, ITZ and outer surface porosity of carbonated and non-carbonated RCA's. The
287 microscope is operated at 20 kV acceleration voltage and 5.7 – 6.7 mm working distance.
288 Backscattered electron images as well as point and line EDS full element analyses are carried out with
289 the following objectives: (1) study the effects of carbonation on ITZ, (2) observe changes in surface
290 porosity, and (3) explore the effects of carbonation on unhydrated cement grains.

3 Results and discussion

The chosen sequence of crushing steps is found to result in RCA with a particle size distribution close to those of starting 0/3 siliceous sand and 7/14 limestone. Figure 2 shows the sieve analysis results of the >3 mm and <3 mm fractions of RCA in comparison with the 0/3 and 7/14 aggregates. It is observed that the coarse RCA has a particle size distribution close to that of 7/14 limestone. Maintaining similar particle size is important for obtaining comparable water absorption results. This is because it eliminates the particle size effect and helps determine (1) how much change in the physical properties will take place due to the presence of hydrated cement paste on RCA, and (2) how much change in such properties a given carbonation procedure can result in. It is also important to measure the cement (draw powder) and mortar (cement + water + sand) contents of the RCA as it affects the water absorption and the CO₂ uptake. The coarse RCA contains average cement (dry powder) and mortar (cement + bound water + sand) contents of 11.8%, and 44.9%, respectively. It is also worth noting that the average cement content in the fine fraction was found to be 25.0%. Table 5 shows the density, water absorption, and porosity results of RCA carbonated under different conditions. A more detailed discussion on each parameter is provided in separate sections as follows.

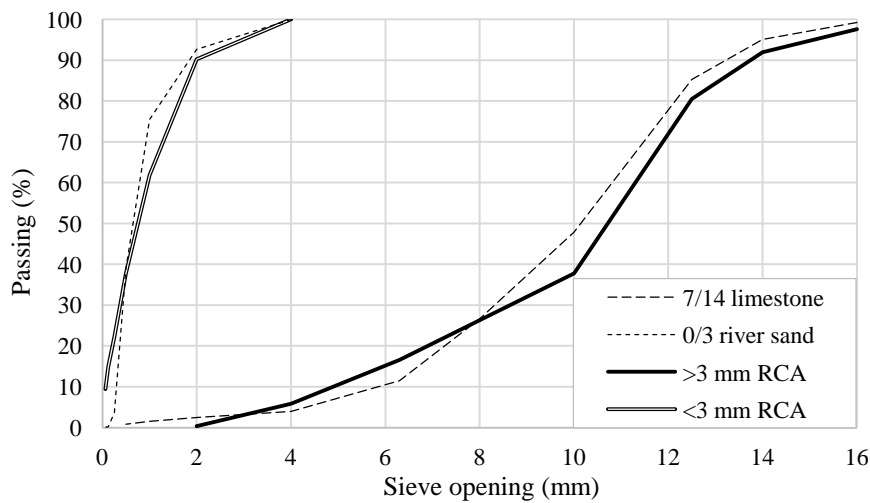


Figure 2. Sieve analysis results of RCA in comparison with the fresh aggregates.

Table 5. The results of RCA density, water absorption and porosity before after carbonation.

Exp. no.	Label	Apparent particle density (kg/m ³)	Oven-dried density (kg/m ³)	Water absorption (%)	Porosity (%)
1	CTRL	2663	2335	5.26	14.04
2	W/B/20C/1b/1h	2687	2336	5.58	15.01
3	W/B/20C/5b/1h	2678	2350	5.19	13.94
4	W/B/60C/1b/1h	2683	2334	5.56	14.95
5	W/B/60C/5b/1h	2679	2335	5.48	14.71

6	W/NB/20C/1b/4h	2676	2322	5.7	15.27
7	W/NB/20C/5b/4h	2688	2344	5.45	14.68
8	W/NB/20C/1b/24h	2687	2308	6.09	16.4
9	PC/B/20C/1b/1h	2678	2343	5.34	14.31
10	PC/B/20C/5b/1h	2677	2338	5.41	14.51
11	PC/NB/20C/1b/1h	2678	2360	5.03	13.49
12	PC/NB/20C/1b/24h	2689	2420	4.11	11.10
13	PC/NB/20C/5b/1h	2670	2393	4.32	11.57
14	PC/NB/20C/5b/4h	2678	2407	4.21	11.27
15	AD/NB/20C/5b/4h	2662	2417	3.82	10.15
16	OD/NB/20C/5b/4h	2674	2422	3.88	10.40

Water absorption results

It is observed in Table 5 that carbonation of RCA under “Wet” or “Boosted” humidity conditions leads to no improvement in water absorption (see water absorption and porosity results of Exp.’s 1 to 10). Comparing the water absorption of the control RCA with the first nine cases where either the RCA is “Wet” or the relative humidity is “Boosted”, reveals that the water absorption capacity may only grow larger after carbonation. This could be attributed to the fact that soaking RCA in water one week prior to testing for water absorption causes (visible) leaching of portlandite from the adhered cement paste, which in turn results in an increase in RCA porosity and thus water absorption. During the carbonation process itself, some extra water is produced due to the conversion of portlandite to calcium carbonate and water. When excessive free water is already available on the RCA surface (due to soaking), carbonation is found to cause further portlandite leaching. This is visually confirmed by the occasional formation of milky water ponds at the bottom of the carbonation chamber (as a result of dripping). It could be concluded that under such conditions (i.e., “Wet” or “Boosted”), variations in CO₂ pressure, carbonation temperature and duration make no meaningful improvement in the carbonation outcome. In fact, a comparison of the NP/W/NB/20C/1b RCA carbonated for 1, 4 and 24 h (Exp.’s 2, 6 and 8) reveals that prolonging carbonation of wet RCA can only lead to a continuous increase in water absorption and porosity (see Table 5). A more detailed discussion is provided after microstructural investigations.

It is prudent to also take into account the inherent variability of the results for successful detection of meaningful variations in water absorption. Figure 3 shows the relative drops in water absorption of RCA in comparison with the control (in percentage points). The error bars show the simultaneous confidence intervals of such relative drops which are constructed such that only 10% chance is left for error (i.e., the event where in one or more cases, the true value of drop does not fall into the provided interval). At first glance, this figure confirms the general increase in water absorption after carbonation under “Boosted” or “Wet” conditions. The error bars, however, suggest that the increases are not significant in most cases. It can be demonstrated that if the error bar of the difference in water

338 absorption of two RCA encompasses zero, such difference is not statistically significant. Therefore,
339 the only two instances where the increase in water absorption is meaningful are the cases of “Wet”
340 RCA carbonated under 1 bar CO₂ pressure at 20 °C for 4 and 24 h (see Exp.’s 6 and 8).

341 However, when it comes to the cases where neither the RCA is “Wet” nor the chamber relative
342 humidity is “Boosted”, promising results surface out and the roles of other carbonation parameters
343 mentioned earlier become clearer (see Exp.’s 11 to 16). While carbonation of “Preconditioned” RCA
344 under 1 bar at 20 °C for 1 h leads to a minimal drop in water absorption (Exp. 11), extension of the
345 carbonation duration to 24 h results in a 1.15% absolute decrease in water absorption which translates
346 to nearly 22% relative drop in such parameter (notice that carbonation of “Wet” RCA under similar
347 conditions leads to 15.8% relative inflation in water absorption).

348 Increase in CO₂ pressure to 5 bars for a 1-h or 4-h carbonation time leads to 0.94% and 1.05%
349 absolute decrease in water absorption values (see Exp.’s 13 and 14), which is in agreement with the
350 results previously published by Xuan et al. [27] where they reported 1.15% drop in water absorption
351 after carbonation of 5 – 20 mm RCA at 5 bars for 24 h. Since carbonation at 5 bars leads to results
352 similar to those of the case of 1 bar in much shorter time, carbonation at 5 bars for 4 h is adopted for
353 the next experiments. Further elimination of RCA moisture by drying at 50% RH (i.e., “Air-dried”)
354 was found to result in additional drop in water absorption and porosity (see Exp. 15). Complete
355 elimination of aggregate moisture (i.e., “Oven-dried”) is also more effective than carbonation of RCA
356 preconditioned at 95% RH, but not as effective as that of “Air-dried” RCA (compare Exp.’s 15 and
357 16). The highest relative drop in water absorption was recorded for the case of “Air-dried” RCA
358 carbonated under “Not-boosted” RH and 5 bar CO₂ pressure at 20 °C for 4 h (27.4%). Zhang et al.
359 [38] also reported 23-28% drop in such parameter after carbonating aggregates at 20 °C, 60% RH and
360 20% CO₂ concentration (results varied depending on aggregate type).

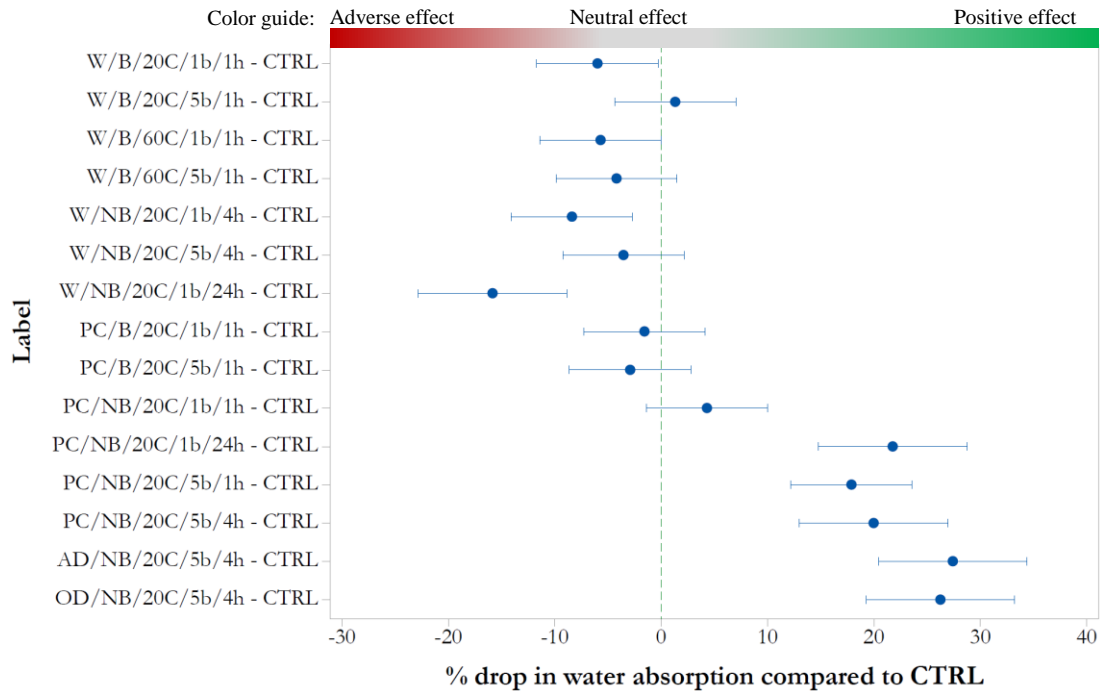


Figure 3. Relative drops in water absorption of RCA compared to the control along with Dunnett Simultaneous 90% confidence intervals (if an interval does not contain zero, the corresponding drop in water absorption is statistically significant).

Porosity results

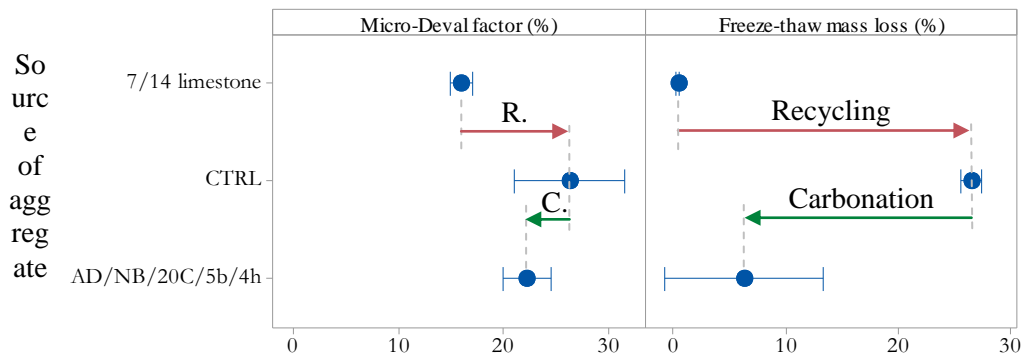
Using Eq. (1), the values of porosity for the fresh materials and RCA are reported in Table 2 and Table 5, respectively. Notice how the porosity of similar size aggregate increases from its original value of 1.73% to an average value of 14.04% after recycling (over 7 times increase from the original value). As shown in Table 5, carbonation of RCA under “Wet” or “Boosted” conditions leads to occasional increase in porosity, which as discussed earlier is probably due to leaching of portlandite during soaking and testing for water absorption. However, carbonation under “Preconditioned” and “Not boosted” conditions for extended durations of time results in considerable drop in porosity (20.9% decrease in the case of PC/NB/20C/1b/24h and 25.3% in the case of PC/NB/20C/1b/24h). However, similar to the case of water absorption, the highest drop in porosity was observed in the case of “Air-dried” RCA (i.e., Exp. 15: AD/NB/20C/5b/4h with 25.9% relative drop in porosity). The obtained values are larger than previously reported by Zhao et al. [31] (7.8 – 14.1%).

Micro-Deval and freeze-thaw resistance results

The starting limestone aggregate showed no more than $16.0 \pm 0.52\%$ mass wear-out in the micro-Deval test while that of CTRL was found to be $26.3 \pm 2.62\%$. The RCA carbonated under optimal

382 conditions (AD/NB/20C/1b/24h) showed an MDE factor of $22.2 \pm 1.13\%$, which is $39 \pm 1.8\%$ higher
 383 than that of fresh aggregates. However, compared to CTRL there has been approximately 40%
 384 regression of the MDE factor (to its original value) as a result of carbonation (see Figure 4).

385 A similar trend has been found in the freeze–thaw resistance results. While the freeze–thaw resistance
 386 of the fresh limestone aggregates was no more than $0.3 \pm 0.12\%$, the recycled concrete showed $26.5 \pm$
 387 0.64% degradation during the freezing and thawing cycles. Carbonation under the said optimal
 388 conditions could significantly reduce this parameter to $8.8 \pm 2.10\%$ (i.e., 77% regression).



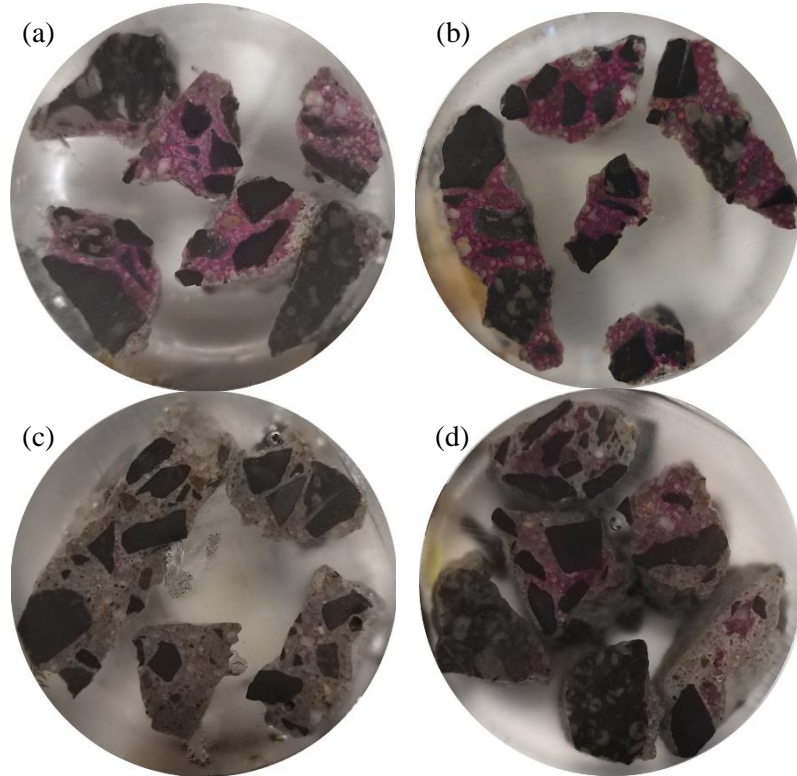
391 Figure 4. The Freeze–thaw resistance and micro–Deval result of fresh, recycled and carbonated
 392 aggregates.

394 Depth of carbonation results

395 The RCA samples chosen for studying the depth of carbonation are selected based on their water
 396 absorption results and also such that they are easily comparable. The selected samples are as follows:
 397 CTRL; W/NB/20C/1b/24h; PC/NB/20C/1b/24h; PC/NB/20C/1b/24h; and AD/NB/20C/5b/4h. Figure
 398 5 shows the response of the cut RCA grains to exposure to the phenolphthalein indicator solution.

399 As expected, the CTRL specimen (i.e., non–carbonated RCA) completely turned pink in the cement
 400 matrix regions minutes after exposure (see Figure 5a). Note that the non–pink regions are either
 401 composed of limestone aggregates or they are covered with the epoxy. The RCA carbonated under
 402 “Wet” conditions under 1 bar CO₂ pressure at 20 °C for 24 hour (Figure 5b) also completely turned
 403 pink indicating incomplete/insignificant carbonation. Same RCA carbonated under similar conditions
 404 but after being preconditioned at 95% RH (Figure 5c) showed almost no color change (except for the
 405 very small regions at the centers of large grains). This suggests that if the aggregates are fully
 406 saturated during carbonation, the rate of CO₂ diffusion in the RCA will considerably decrease. The
 407 water absorption results also reveal inefficiency of carbonation under wet conditions. The BSE

408 images later shown and discussed in Figure 6 on the wet-carbonated RCA also suggest no surface
409 densification.



411
412 Figure 5. The cut RCA grains exposed to phenolphthalein indicator solution: (a) CTRL (non-
413 carbonated RCA); (b) W/NB/20C/1b/24h; (c) PC/NB/20C/1b/24h; (d) AD/NB/20C/5b/4h.

414
415 Interestingly enough, the RCA carbonated under “Air-dried” conditions at 5 bars for 4 h showed a
416 considerable color change (see Figure 5d). It is observed that some grains did not show any color
417 change up to a few millimeters in depth while others have completely turned pink, which implies an
418 incomplete and non-uniform carbonation. However, as explained earlier the carbonation procedure
419 applied to this RCA resulted in the highest drop in water absorption. This can be reconciled by
420 noticing the fact that phenolphthalein turns pink at pH above 10. Therefore, if pore solution pH drops
421 from its original value (typically 13.6–13.8 [39]) to any value above 10 (due to carbonation), the
422 phenolphthalein will still indicate a basic solution. As such, a pink color should not be interpreted as a
423 sign of no carbonation, because the pore solution pH can remain above 10 even after full carbonation
424 of portlandite and partial carbonation of C–S–H [40]. In the case of AD/NB/20C/5b/4h, it appears that
425 despite the high CO₂ pressure, C–S–H has not sufficiently been carbonated due to the short
426 carbonation time (4 h), while the lowest porosity among all carbonated RCA’s is observed in this
427 case. This suggests that a higher degree of carbonation does not necessarily translate to a higher drop

in RCA surface porosity. A more in-depth investigation of the microstructure of the same samples is thus presented in the next section.

Microstructural investigation results

Figure 6 shows the BSE images of the selected RCA samples. Comparing CTRL (Figure 6a) and the RCA carbonated under “Wet” conditions (Figure 6b) with the latter two RCA’s (i.e., Figure 6c and d which experienced effective carbonations), it is evident that carbonation transforms the transition zone between the aggregates and the old cement paste. While portlandite deposits are clearly visible in the ITZ regions of CTRL and W/NB/20C/1b/24h (Figure 6b), no such phase is observable in the latter two cases.

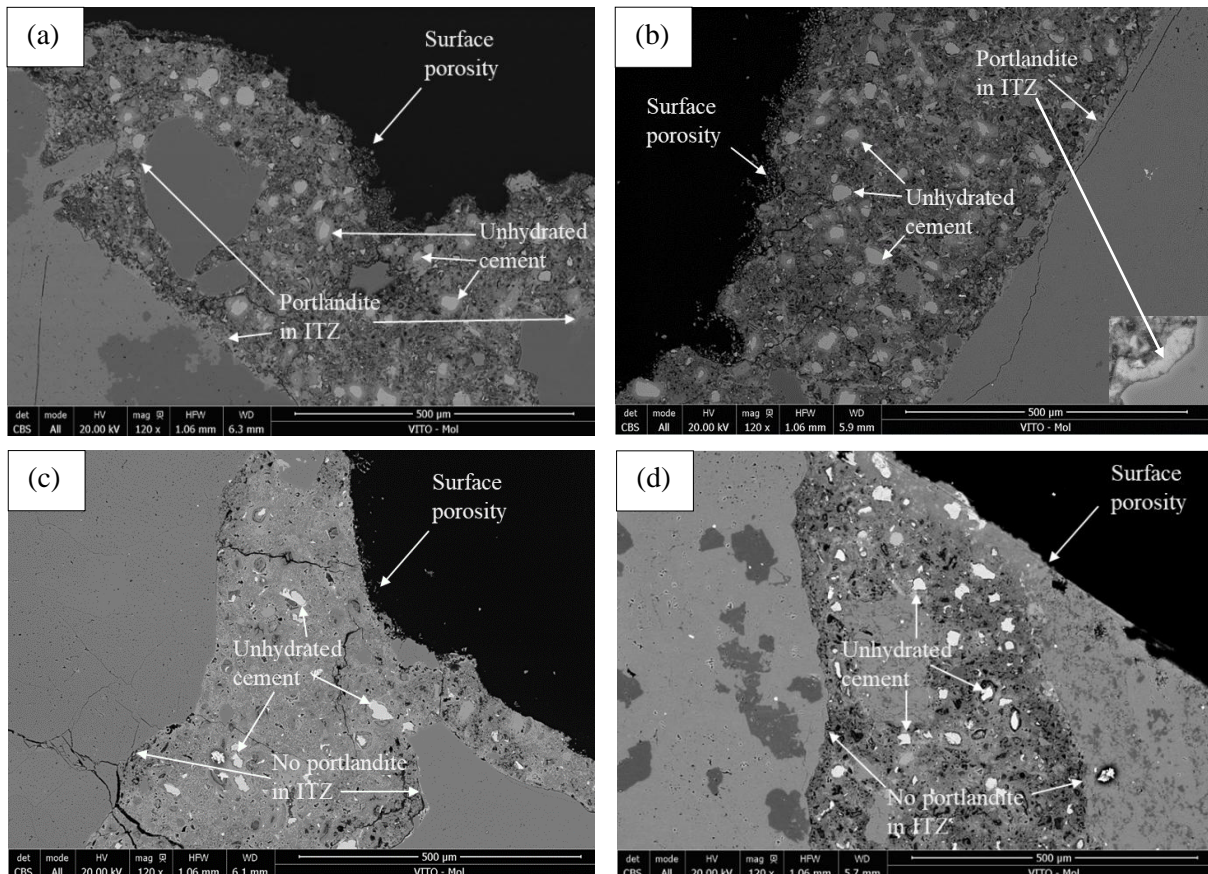


Figure 6. The BSE images of RCA grains: (a) CTRL (non-carbonated RCA); (b) W/NB/20C/1b/24h; (c) PC/NB/20C/1b/24h; (d) AD/NB/20C/5b/4h.

In addition, carbonation results in occasional formation of a dark rim around the unhydrated cement grains which is the result of decalcification of the reacted (C–S–H) rim during carbonation (which can turn into silica gel upon full carbonation). Such transformation has been previously reported and

446 theorized by Mu et al. (2018) [41]. Moreover, carbonation of β - and γ - C_2S has been investigated in
1 447 the past by numerous researchers [42,43,44,45]. It is commonly concluded that while the hydration of
2 448 C_2S is rather slow, it can undergo carbonation very rapidly, sequester significant amounts of CO_2 , and
3 449 undergo large volumetric increase (51.2% and 172%, respectively). As carbonation liberates water
4 450 from cement hydrates, it may also lead to reactivation or renewed hydration of the residual cement
5 451 grains.

10 452 Moreover, carbonation under “Not-boosted” RH conditions results in considerable densification of the
11 453 surface (compare the surface porosity of RCA grains in Figure 6a and b with Figure 6c and d). The
12 454 formation of such densified rim on the exterior parts of the cement paste in carbonated RCA grains
13 455 could be indicative of the outward aqueous transport of Ca from the carbonating interior. Two
14 456 experimental observations would support this claim: 1) during carbonation of RCA under saturated
15 457 RH, it was observed that the RCA released noticeable amounts of moisture, which often turned into
16 458 run-off water and, at the end of carbonation process, was visually detected at the bottom of the
17 459 carbonation reactor. Its milky appearance suggested the presence of suspended calcium carbonate
18 460 precipitates that leached out from the RCAs during carbonation while being exposed to excessive
19 461 humidity, 2) since carbonation is expected to eventually result in an increase in porosity, the outer
20 462 RCA surface would be expected to be more porous than the interior (due to more exposure).
21 463 However, judging by Figure 6c and d, it could be argued that the opposite is observed for RCA
22 464 carbonated under optimal conditions. Combined, both observations suggest that the outward reactive
23 465 transport of Ca-rich pore solution is the most plausible densification mechanism. Depending on the
24 466 reactor RH, two contrasting outcomes can be realized: 1) in case of saturated reactor RH, the RCA
25 467 moisture accumulates at the RCA surface until liquid run-off (drip-off) from the surface occurs.
26 468 Dissolved ions are carried along with the liquid resulting in Ca-leaching and a porosity increase, as
27 469 indeed observed for carbonation of wet RCA in this study (the mechanism is sketched in Figure 7a).
28 470 In case of low reactor RH, the driven-out moisture is efficiently evaporated at the RCA surface and
29 471 the accumulating Ca would precipitate as $CaCO_3$ at the surface, thus causing densification of the outer
30 472 RCA surface (see Figure 7b).

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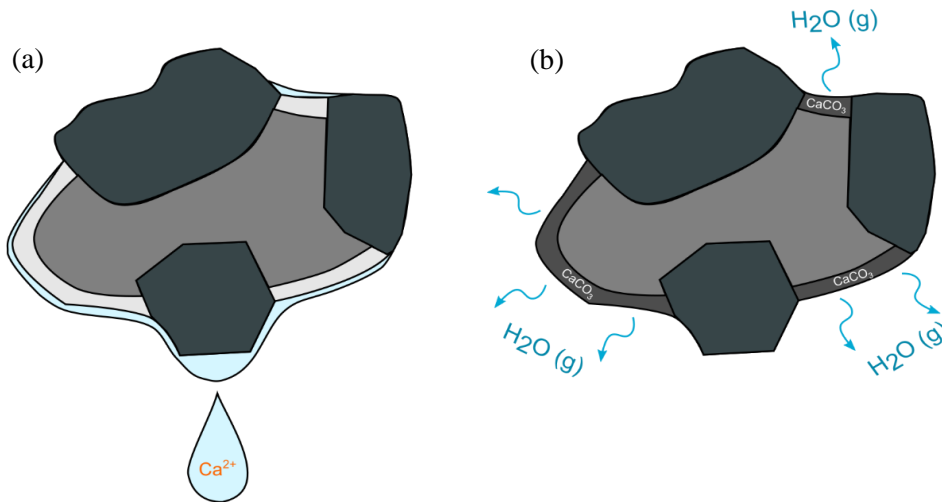


Figure 7. The proposed outward Ca-transport mechanism explains a) leaching and porosity increase at saturated reactor RH, and b) formation of a densified surface rim at low reactor RH.

Cost/benefit assessment of RCA carbonation

Mineral carbonation is still in its infancy and its application is limited due to its relatively high and varying cost [46]. With respect to RCA, however, it is considered as a rapid, cost effective and efficient method for improving the properties of RCA when compared to other common methods [26,27]. Given ~11.8% cement content per each tonne of coarse RCA and the maximum CO₂ uptake capacity observed by the researchers (~40%), it can be concluded that approximately 45–50 kg of CO₂ will be absorbed by each tonne of RCA.

4 Conclusions

The following conclusions can be drawn from the presented research:

- Carbonation of recycled concrete aggregate (RCA) under wet conditions or moist atmosphere does not lead to any improvement in its water absorption, porosity or microstructure regardless of CO₂ pressure, temperature.
- Carbonation of RCA preconditioned at ≤95% relative humidity, under medium to high levels of CO₂ pressure for prolonged durations of time (e.g., 1 bar for 24 hours or 5 bars for 4 hours) in the absence of an external source of water vapor leads to considerable refinement of the pore structure. Up to 27% relative drop in water absorption and porosity, 40% regression of micro-Deval resistance and 77% in freeze-thaw resistance was recorded for air-dried RCA carbonated under 5 bars CO₂ pressure at 20 °C for 4 hours. It can thus be concluded that carbonation of RCA prior to use in concrete can lead to considerable drop in its water absorption and porosity (and thus improvement of its durability against cycles of freezing and thawing) and an increase in its mechanical properties.

- 499 - The visual and microstructural investigations suggest that carbonation of RCA under the
1 500 above proper conditions (i.e., RCA preconditioned at 95% RH and absence of an external
2 501 source of moisture) has the following sequential effects: 1) continuous carbonation of
3 502 portlandite and C-S-H, 2) generation of extra water as per portlandite carbonation reaction, 3)
4 503 transportation of pore solution and dissolved Ca^{2+} to the exterior surface of the grains, 4)
5 504 effective evaporation of water at the surface while maintaining an air-accessible pore
6 505 structure, and 5) further carbonation of calcium and deposition at the surface. This was found
7 506 to result in the densification of the RCA surface, reduction of surface porosity and increase in
8 507 RCA resistance against wear, and cycles of freezing and thawing. However, carbonation of
9 508 RCA under wet conditions and an excess of ambient moisture for prolonged durations of time
10 509 (e.g., 24 hours) was found to result in an increase in porosity due to the leaching of
11 510 portlandite from the RCA as carbonation continued.
12 511 - Successful carbonation was found to transform the ITZ and form a dark rim around the
13 512 unhydrated alite and belite grains present in the cement matrix indicating hydraulic
14 513 reactivation and carbonation of the unhydrated grains.
15 514

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