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1 Carbonated recycled concrete aggregates in construction: potential
2 and bottlenecks identified by RILEM TC 309-MCP

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36

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66 **Abstract**

67 This review by Working Group 1 of the RILEM TC 309-MCP discusses recent advances in the beneficial
68 carbonation treatment of recycled concrete aggregates (RCA). The impact of carbonation on RCA
69 properties as well as the microstructure and performance of concrete and other construction
70 materials made thereof is critically reviewed. The increasing focus on environmentally friendly
71 building practices has led to a greater interest in the CO₂ uptake associated with carbonation
72 processing. Furthermore, emphasis is placed on the importance of adopting tailored strategies to
73 optimise the carbonation process based on the quality and type of RCA. Evidence in the literature
74 highlights the beneficiation potential of carbonation processing in improving RCA properties and
75 performance, which translates in variable degrees of enhancement of the performance of concrete or
76 other applications made thereof. The review concludes that, to date, significant techno-economic
77 challenges remain to be addressed to improve the competitiveness of the technology, notably in terms
78 of upscaling and refining life cycle assessment data.

79 **1. Introduction**

80 Enforced carbonation of recycled concrete aggregate (RCA) is a growing technology with enormous
81 potential. This carbonation treatment uses CO₂ to improve the properties of cement-bearing RCA and
82 make it more suitable for use in construction applications. While carbonation occurs naturally over
83 the course of years to decades, carbonated RCA (cRCA) for construction is intentionally produced by
84 tailored processes, intensifying the treatment over several hours or days. During the carbonation

85 process, RCA is subjected to increased CO₂ concentration, which expedites the precipitation of the
86 carbonatable phases as solid carbonates.

87 Intensive research into cRCA has identified and optimised the main parameters that control the
88 process. These specific values for the parameters depend on whether the target is to enhance RCA
89 properties or maximise CO₂ uptake, which are the two main advantages of RCA carbonation. Initially,
90 most attention was directed to the property enhancement of coarser RCA fractions (>2-4 mm). Fine
91 fractions (<2-4 mm) were overlooked due to their more restricted market acceptance and use in
92 concrete production (1, 2). The interest in fine fractions progressively grew as enforced carbonation
93 gained relevance in terms of its carbon capture and utilization (CCU) (3-5). While early studies
94 indicated that CO₂ uptake of RCA was of the order of 5-11 kg/ton of RCA (6, 7), with a surprisingly
95 unmarked contrast between natural carbonation and enforced carbonation (even at 5 bar pressure),
96 more recent research (8) reports more promising values of 10-50 kg/ton of RCA. Recycled concrete
97 fines have a higher content of carbonatable phases (9) and can carbonate faster than the coarse
98 fraction, resulting in greater CO₂ uptake potential and improved properties that enable easier use in
99 concrete. Consequently, there is significant interest in carbonation treatment, regardless of the
100 particle size of RCA.

101 Through subsequent validation of the technology, extensive research into the effects of cRCA on
102 concrete performance has also been conducted. These findings pertain mainly to coarse RCA fractions,
103 while only in recent years have fine fractions started to receive comparable attention.

104 Beyond concrete applications, cRCA can serve various other applications, e.g., screed, mortar, road
105 construction, and soil stabilisation. The low price of aggregates for these applications can hardly make
106 cRCA sufficiently competitive. However, carbonation can add value due to the CO₂ uptake itself (8).
107 The CO₂ capture can be monetized when coming from industrial emitters through mechanisms such
108 as ETS credits.

109 Despite the extensive research, cRCA has yet to gain substantial market uptake. Most of the pending
110 work lies in upscaling, demonstration, and economic feasibility studies, which are uncommon in
111 existing literature (10, 11). Refining production costs and deriving benefits from carbon pricing may
112 contribute in this regard. Barriers are not limited to economical feasibility, but also cultural obstacles
113 have slowed down implementation in some cases (12) (including lack of awareness, rigid production
114 methods, and restrictive regulation).

115 This paper offers a critical review of recent advances in enforced carbonation of RCA, addressing
116 processing developments, aggregate property modification, and effects on concrete performance and
117 other types of construction applications. In addition, specific attention is directed to practical
118 implementation aspects in terms of upscaling, cost-benefit analysis, and life cycle analysis. On this
119 basis, pending research needs and general guidelines for using cRCA in construction applications are
120 provided.

121 2. Advances in RCA carbonation processing

122 The main process parameters for optimising the carbonation treatment of RCA include temperature,
123 water content, CO₂ concentration, pressure, duration of exposure, and particle size fraction. These
124 factors vary depending on the method of carbonation, which can be broadly divided into moist or
125 aqueous carbonation (13) (Figure 1). Many examples of optimized parameters for the various enforced
126 carbonation methods are compiled in the annex with Supplementary Information. In moist (or semi-
127 dry) carbonation, static, dynamic, or rolling methods (14) allow continuous contact of a CO₂-enriched
128 gas, though other process parameters like pressure, temperature, relative humidity (moisture in the

129 air or carbonation environment), and moisture content (water content in the sample), can still be
130 tuned to achieve better results. For example, typical ranges of parameters adopted for a carbonation
131 treatment are $(20\pm 2)\%$ CO₂ concentration, $(60\pm 5)\%$ RH, and (20 ± 2) °C temperature (15, 16). In aqueous
132 carbonation, the liquid-solid phase interaction is promoted (Figure 1, c), facilitating the reaction
133 between hydration products, CO₂, and water. This method claims to be more efficient than moist
134 carbonation in terms of the carbonation conversion rate or CO₂ uptake. These optimal conditions for
135 maximum CO₂ uptake do not necessarily match those for optimal RCA property enhancement (17-21).
136 In aqueous carbonation, there is a possibility that the carbonation products can precipitate in the
137 solution and render the residual aggregate more porous. For example, it is reported that floating
138 process conditions during carbonation can lead to suboptimal results and diminish the desired effects
139 (15, 16). Nevertheless, recycled concrete fines would be the most suitable candidate for the aqueous
140 carbonation method owing to the presence of more hydrated phases on the surface (18).

141 The pretreatment of the RCA greatly impacts the efficiency of the carbonation treatment. In most
142 cases, semi-dried RCA is used in cases of semi-dry carbonation, whereas RCA is soaked in water for
143 aqueous carbonation scenarios. Previous studies indicated that appropriate drying decreased the
144 saturation degree of the RCA porosity and hence promoted the diffusion of CO₂ through the gas phase
145 (22, 23). Similarly, aqueous carbonation may improve with pretreatment using magnesium or calcium
146 solutions instead of water soaking (17, 24, 25).

147 Other than the carbonation methods and pretreatment conditions, influential parameters (e.g.,
148 pressure, temperature, relative humidity, moisture content, CO₂ concentration, duration of exposure),
149 and their interaction effects are to be considered to attain the maximum efficiency of the carbonation
150 treatment of RCA. The size of the RCA has a great impact on the CO₂ uptake capacity during both moist
151 and aqueous carbonation (18, 23, 26), though greater influence is observed in the latter method
152 (Figure 2, a). The finer the RCA, the higher the CO₂ uptake. Fine RCA usually contains a higher hardened
153 cement paste content, hence a higher carbonation potential. A larger specific surface area with
154 decreasing aggregate size is an additional benefit for enhanced reactivity. Despite the natural
155 carbonation that occurs during storage, the CO₂ uptake by RCA piles is low compared to subsequent
156 enforced carbonation, and it only reaches the most superficial layers of the stockpiles (27).

157 In moist carbonation, there is an “optimum effect” considering the influence of relative humidity
158 (Figure 2, b) (28). Most of the studies adopted a relative humidity value in the range of 50 to 70% as
159 being optimal (see examples in the annex with Supplementary Information). This phenomenon is
160 related to the facilitation of the transportation of CO₂ into the RCA pore structure. Intermediate
161 moisture levels provide transport pathways through the non-saturated porosity, while at the same
162 time enabling the dissolution of CO₂ and hydration products in moisture-filled fine porosity, thereby
163 accelerating the carbonation process. In contrast, water lacks as a dissolution and reaction medium at
164 very low moisture levels, while excessive moisture would hinder the gas-phase diffusion of CO₂, and
165 hence the CO₂ uptake of RCAs is reduced or limited to the exposed surface (28, 29). The issue of
166 maintaining optimal internal saturation levels does not play a role in the aqueous carbonation
167 methods (Figure 2, c). In aqueous carbonation, the CO₂ is already dissolved in the water, so the gas-
168 phase diffusion process is not involved. On the other hand, in aqueous carbonation Ca and Mg may
169 leach from the RCA and precipitate in the aqueous reaction medium, hence increasing the porosity of
170 the RCA and deteriorating its performance.

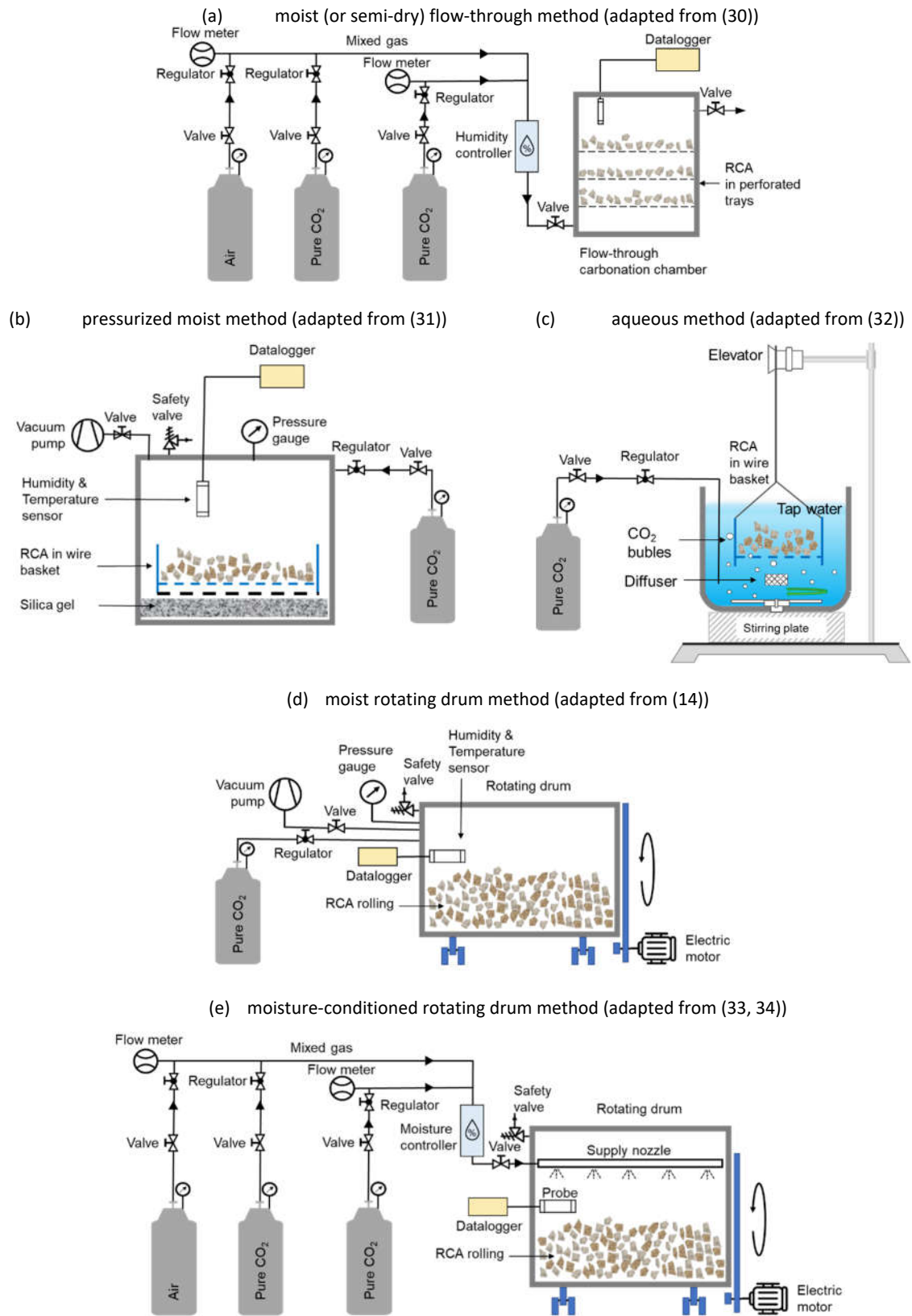
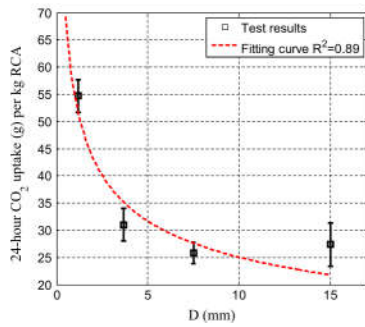
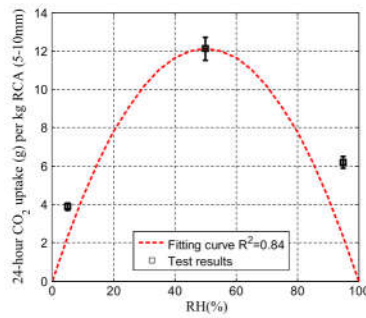


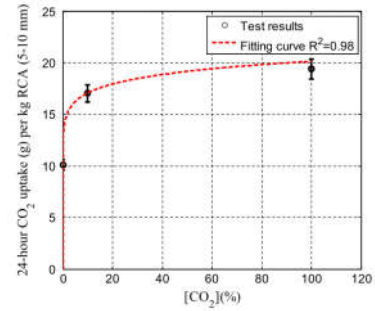
Figure 1. Methods of carbonation.



(a) Size effect



(b) RH



(c) CO₂ concentration

Figure 2. Influence of different parameters on the CO₂ uptake potential (reproduced from (28)).

Temperature increases carbonation reaction rates yet decreases CO₂ solubility in water. Temperature also influences the formation of different polymorphs of calcium carbonate (35). While calcite is the most commonly formed and most stable polymorph, a combination of a low temperature (20±5°C, room temperature) and a high CO₂ concentration (>10%) can result in aragonite formation. At high temperatures (70±5 °C), faster carbonation (12 hours) results in vaterite, whereas with an increase in carbonation time to 24 hours, more stable calcite is formed. Considering the CO₂ concentration for the treatment of RCA, 20% is considered the preferable input in most of the cases (22, 23, 35, 36). There is no substantial improvement observed at high concentrations of CO₂ (Figure 2, c). This behaviour at high CO₂ concentrations is attributed to the formation of solid carbonation products on the RCA surface and pore space, hindering further diffusion of CO₂ (37). However, enforced carbonation with a higher CO₂ concentration is reported to be beneficial in pressurized and/or aqueous conditions (26, 28). Higher CO₂ concentrations coupled with pressurized conditions could help in further diffusion and thereby increase CO₂ uptake to a certain extent. For example, pressurized methods could give almost twice the sequestration compared to flow through (20 g/kg vs. 12 g/kg) (28).

The CO₂ uptake of RCAs increases with the duration of carbonation. After a certain period of time, the pores will be filled with carbonation products, and the path for diffusion will be blocked. Generally, 24-72 h of pressurized carbonation is sufficient for moist carbonation, while it only needs 6 h for aqueous carbonation (22, 32). Still, long-term stability is assured in moist pressurized carbonation, as the liquid-solid method results in dissolution of the products at later stages (21). Considering the moist carbonation process, the pH of the system is identified as an important factor. High pH (alkaline) increases the solubility of CO₂ and the precipitation of carbonated products. On the other hand, low pH (acidic) could accelerate the dissolution of Ca and Mg from the carbonatable phases but is not beneficial for the precipitation of carbonates. Given the important Ca concentration of cement, prohibitively large amounts of acid would be required to reach a low pH, rendering pH-swing approaches impractical (38).

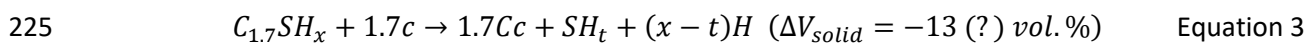
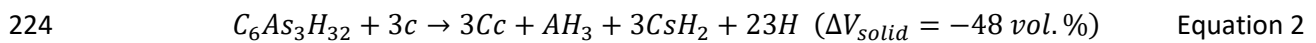
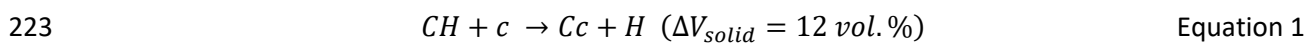
3. Property modification of cRCA

Carbonation has shown promising potential for enhancing certain RCA properties relevant to the production of new concrete (39-41). The influence of various RCA material and carbonation processing parameters on the physical properties of RCA has been a subject of significant research efforts. This section first presents an overview of the underlying chemical reactions, followed by a comprehensive discussion of the modifications in physical properties and microstructure.

3.1. Chemical changes

In RCA, carbonation involves the chemical reaction of CO₂ with calcium-rich cement hydrates. This reaction primarily leads to the formation of calcium carbonate and an array of residual phases

210 comprising silica, alumina, and sulfate. Among cement hydrates, calcium hydroxide (CH) exhibits the
 211 highest susceptibility to carbonation, followed by C-S-H and, eventually, ettringite, and AFm hydrates.
 212 Hydrotalcite appears not to be susceptible to carbonation (42, 43). As hydrates are converted into
 213 carbonates, bound water is released while CO₂ is taken up. Reported CO₂ uptake for RCA carbonation
 214 varies considerably, spanning from approximately 5 to 50 kilograms of CO₂ per tonne of RCA (8, 15,
 215 16, 28, 44, 45). This broad range reflects dependencies on various factors, including particle size (15,
 216 16, 28) and ageing (44) but also recycling and carbonation process parameters (10, 20, 32). The solid
 217 volume change brought about by carbonation is contingent upon the individual reactions involved.
 218 For instance, the carbonation of calcium hydroxide (CH) to calcite unequivocally leads to an increase
 219 in solid volume of about 12%. However, as shown in Equations 1-3, for other conversions the solid
 220 volume change would not always be positive. In the case of ettringite, carbonation results in a
 221 substantial reduction in solid volume, while the outcome for C-S-H remains unsettled due to the yet
 222 inconclusive understanding of the density of the resulting silica gel product (46).



226 The equations above adopt phase properties according to (47) and consider the formation of calcite
 227 (molar volume of 36.9 cm³/mol). Most authors confirm calcite as main RCA carbonate product;
 228 however, for carbonation at higher temperatures and lower relative humidities, also vaterite (molar
 229 volume of 37.7 cm³/mol) and, more rarely, aragonite (with a molar volume of 34.2 cm³/mol) have
 230 been reported (48). The type of carbonate polymorphs formed has minor yet noticeable effects on
 231 the overall change in solid volume (41). The implication of the above is that achieving maximum CO₂
 232 uptake does not necessarily align with optimal enhancement of RCA properties. In fact, minimal
 233 porosity and optimal property improvements may be reached at lower carbonation degrees.

234 3.2. Physical properties

235 There is growing consensus and evidence that, when implemented under suitable processing
 236 conditions, carbonation effectively enhances the properties of RCA for utilization in new concrete (41).
 237 Particularly, it demonstrates positive impacts on density, porosity, water absorption, and resistance
 238 to crushing or wear (15, 45).

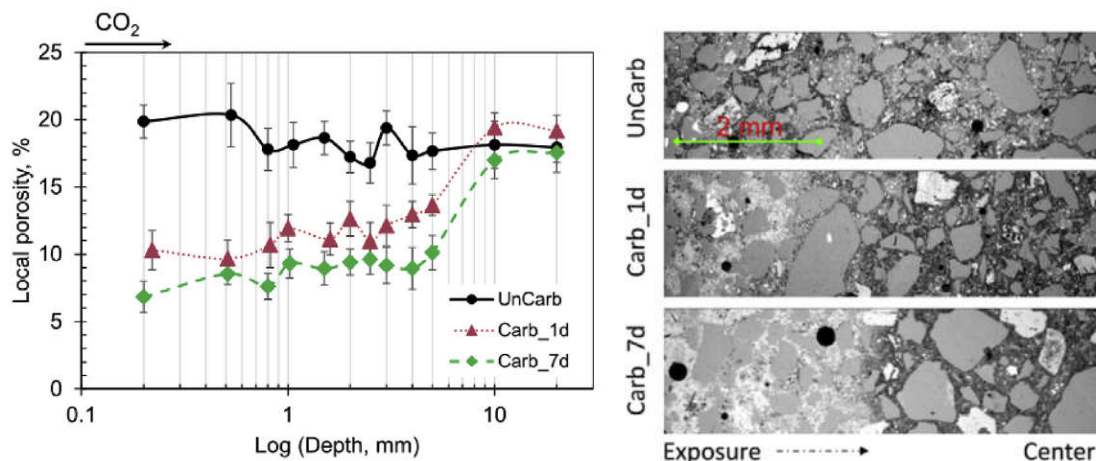
239 Regarding density, carbonation results in an increase in the apparent density of RCA, with the extent
 240 of the increase varying based on factors such as particle size and the characteristics of the parent
 241 concrete (16). The observed range of increase falls between 0.5% and 5% for optimized conditions
 242 (20, 39, 41, 45). Examples of optimized conditions in the literature can be found in the annex with
 243 Supplementary Information. Porosity, closely associated with density, exhibits a consistent reduction
 244 as carbonation progresses. The decrease in porosity aligns with the increase in density and typically
 245 ranges from 20% to 30% (15, 41). RCA water absorption experiences a notable reduction as well, that
 246 range under optimal carbonation conditions (see section 2 for examples) from around 10% to 40% (in
 247 relative terms) or between 1 to 3 wt% (in absolute terms) (7, 24, 45, 49). The extent of reduction can
 248 be more significant for stronger parent concrete and smaller RCA particle sizes. Pre-soaking in lime as
 249 a pre-treatment measure can even lead to reductions of up to 65% (50).

250 In terms of crushing and wear resistance, carbonation contributes to an improvement in properties.
 251 Under optimal conditions, a decrease of approximately 20% to 40% in crushing or wear is observed
 252 (15, 45). On the other hand, carbonation may lead to an increase in RCA stiffness and shrinkage

253 through porosity refinement and drying. This could result in microcracking and eventual RCA property
254 degradation (43).

255 3.3. Microstructural changes

256 Microstructure modifications are at the base of the enhancement of RCA macroscale properties by
257 carbonation. Carbonation, as a reactive transport process, triggers the redistribution of solids within
258 the RCA as CO₂ enters and water is expelled. This process commonly leads to a reduction in porosity
259 at the surface of the RCA due to carbonate precipitation (Figure 3) (49). This reduction in surface
260 porosity leads to a decrease in water absorption and improved crushing resistance. While coarse and
261 mesopores (>50 nm) are filled by carbonation (51), the smallest gel porosity generally tends to
262 increase and coarsen (52). More porous parent concrete tends to carbonate more readily than less
263 porous, stronger concrete. For instance, Thiery et al. (40) reported that C-S-H can be fully carbonated
264 at a w/c ratio of 0.6 and 50% CO₂, while, for a w/c ratio of 0.8, a 10% CO₂ treatment sufficed. Also,
265 other authors found that less porous, higher strength parent concrete translates into cRCA having
266 lower water absorption and higher density (45, 53). This clearly correlates with a higher carbonatable
267 cement content and a lower porosity of the parent concrete. Interestingly, the change in porosity or
268 water absorption is less than can be expected from a mass balance perspective, which is tentatively
269 explained by slower carbonation kinetics and a lower carbonation degree for RCA deriving from
270 higher-strength, denser parent concrete.



271
272 Figure 3. RCA surface porosity reduction by carbonation treatment as measured by electron
273 microscopy (49).

274 4. Effect of RCA carbonation treatment on the interfacial transition zone (ITZ) and 275 concrete performance

276 4.1. The properties of ITZs in the concrete with cRCA

277 The ITZ forming between the aggregates and mortar (54, 55) is crucial to concrete performance. Since
278 RCAs are multiphasic, additional ITZ types (to that forming between new natural aggregates (NA) and
279 new mortar) are present in RCA concrete (31, 56). These include the old ITZ between old NA and old
280 mortar and the new ITZs between old mortar or old NA and new mortar. The carbonation treatment
281 will impact both the extent to which it modifies the pore structure and the texture of the RCA particles.

282 4.1.1. The old ITZ in the cRCA

283 Table 1 summarizes how carbonation affects the micromechanical properties and thickness of the old
284 ITZ within the cRCA. Enforced carbonation increases the microhardness of old ITZ (e.g., by up to 41.7%
285 (57)) and decreases its thickness (e.g., by up to 85% (58)). The relative enhancement in microhardness

286 of old ITZ increases with carbonation time but decreases with the increase in the strength class of the
 287 parent concrete (i.e., the relative improvement of already very dense ITZ is more moderate).
 288 Carbonation pressure, time, and initial moisture affect the average elastic modulus and thickness of
 289 the old ITZ. Higher carbonation pressure and longer duration boost elastic modulus and reduce old ITZ
 290 thickness. The best results (25% thickness reduction to 45 μm and 35% increase in elastic modulus to
 291 33 GPa) were obtained with 12 hours of carbonation at 1 bar. These improvements partially explain
 292 the improved mechanical properties of concrete (59). After 3 h of carbonation at 1 bar pressure,
 293 further improvement in the properties of the old ITZ becomes less significant. The optimal RCA
 294 moisture content to improve the old ITZ was reported in the range of 1.0 to 1.81% (the maximum
 295 improvement at 1.81% decreased ITZ thickness by 16.7% and increased modulus by 29.1%). However,
 296 the opposite trend was observed with a moisture content of 3.0%.

297 Table 1. The micromechanical properties of old ITZ under different carbonation conditions in
 298 moist carbonation method.

Particle size (mm)	Parent material (MPa)	Curing age	Carbonation parameters						Enhancement rate of properties of ITZ (%)			Ref.			
			CC (%)	IMC (%)	CP (bar)	t	RH (%)	T (°C)	Thickn ess	Microha rdness	Modulus				
5-20	33.33	n/a	99.99	1.18	0.5	1 h	n/a	30				-8.3	10.2	(59)	
				1.18	1	1 h						-16.7	18.8		
				1.18	2	1 h						-16.7	24.5		
				1.18	4	1 h						-25.0	26.5		
				1	1	1 h						-8.3	12.2		
				3	1	1 h						0.0	9.0		
				1.18	1	3 h						-16.7	26.9		
				1.18	1	12 h						-25.0	34.7		
5-25	n/a	7d	20±3	n/a	n/a	n/a	70±5	20±2				25	n/a	(60)	
		28d										25			
		140d										17			
10-20												-60.3			
												14 d			-79.5
												28 d			-85.8
20-30	n/a	n/a	20	n/a	n/a	14 d	70±5	20±2				7 d	n/a	n/a	
												7 d			-55.5
												28 d			-76.6
30-40						7 d						-54.2			
						14 d						-75.5			
						28 d						-83.0			
n/a	55.6	n/a	100	n/a	0.1	21 d	n/a	25				11.5	n/a	(31)	
	37.5											34.3			
5-20	n/a	n/a	99.9	n/a	n/a	n/a	55	22				n/a	41.7	n/a	(57)
5-26	30	1d	20±3	n/a	n/a	72 h	70±5	20±2				-7.8	13.9	n/a	(61)
		45d										-4.6	15.1		
		90d										-9.4	15.2		
		1d										-7.7	10.3		
		45d										-4.5	14.7		
60	90d	-3.9	14.5												

299 Note: CC – CO₂ concentration; CP – carbonation pressure; IMC – initial moisture content; t – carbonation duration; RH –
 300 relative humidity; T – temperature.

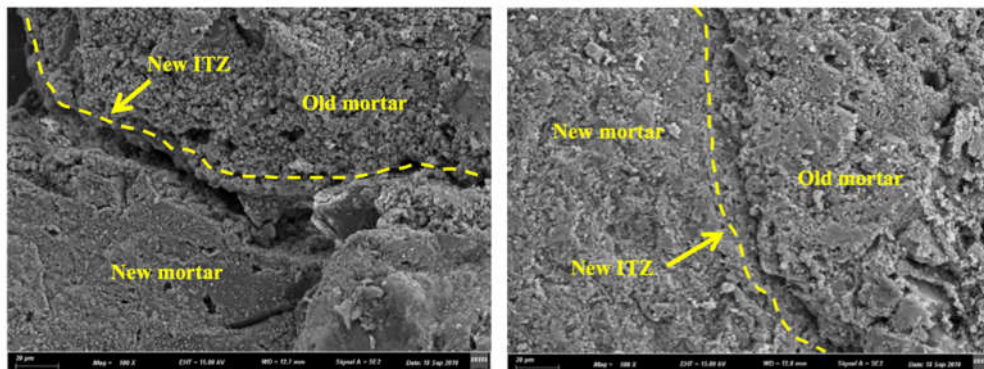
301 4.1.2. The new ITZs between the cRCA and new mortar

302 Enhanced bonding of the cRCA in new concrete is another benefit (62). Notable improvements in
 303 micro-hardness across the new ITZ are reported (39). The analysis is particularly interesting for the ITZ
 304 formed between old mortar and new mortar (i.e., the new ITZ). No significant benefits are to be
 305 expected for the ITZ forming between the old NA and the new mortar (62). Carbonation treatment
 306 only affects old mortar and old ITZ, as these are the phases where the carbonatable phases (cement
 307 hydration products) are found. Overall, calcium carbonates on the RCA surface serve as nucleation
 308 centres for new cement hydrates, while (alumino)silicate carbonation products may participate in

309 pozzolanic reactions with the CH provided by the new cement paste (60). Recent research highlighted
310 that pre-treatment with $Mg(NO_3)_2$ could enhance RCA microstructure properties further by modifying
311 carbonate crystal growth (17).

312 The different carbonation treatment methods may impact differently on the properties of new ITZ
313 (old mortar-new mortar), as revealed by microstructure and micromechanical properties:

314 (1) Moist carbonation: Figure 4 depicts the denser structure with lower porosity and a smaller width
315 of cracks for new ITZ for carbonated old mortar compared to uncarbonated old mortar. After
316 pressurized moist carbonation treatment (Figure 1, b), the improvement is evident, with increased
317 microhardness and decreased thickness of the new ITZ, irrespective of the parent material (similar
318 porosity values were reported for ITZ in concrete with cRCA derived from concrete with $w/c =$
319 0.25 , and respective ITZ with cRCA derived from concrete with $w/c = 0.40$) (62). The carbonation
320 treatment has a more remarkable effect on the immediate regions of the cRCA surface, which
321 reflects the impact of changes in the RCA texture. Statistical analysis connected the new ITZ
322 porosity to the carbonation duration, with a significant decrease in porosity when the carbonation
323 duration was extended from 3 to 7 days (63). The improvement was also reflected in the
324 microhardness of the new ITZ with cRCA, which rose by 24.3% and 51.2%, while the ITZ thickness
325 decreased by 27.3% and 36.4%, respectively, for 3 and 7-days treatments.
326 Similarly, improved microhardness of new ITZ is reported at rates of 13% (60) or 12% (64) for
327 conventional concrete, and 46% (65) for ultra-high-performance concrete by eliminating the
328 interface defects caused by old mortar. Also, ITZ thickness reduction is reported at rates of 43%
329 (64).



330
331 Figure 4. SEM observation of new ITZ formed with RCA and cRCA (64): left: new ITZ for non-
332 carbonated RCA, right: new ITZ for cRCA.

333 (2) Aqueous carbonation: Similar efficiency to improve the elastic modulus of the old mortar was
334 demonstrated for 24 h of moist carbonation and 6 h of aqueous carbonation (21). However, the
335 new ITZ improved more with the 6-hour aqueous carbonation (31% increase in elastic modulus
336 (21), or 36.5% increase in microhardness (20)) compared to the 24-hour pressurized moist
337 carbonation (2% increase in elastic modulus (21), or 17.5% increase in microhardness (20)) and
338 flow gas-solid carbonation (20.4% increase in microhardness (20)). In addition to mortar-mortar
339 ITZ, $CaCO_3$ precipitated on the surface of old NA after aqueous carbonation (18) also has the
340 potential to improve the new ITZ that will form with the new matrix. Further studies are required
341 to validate these observations, as others have reported significant deterioration of aggregate
342 properties upon carbonation in moisture saturated conditions (15).
343 The differences observed in the new ITZ for aqueous and moist carbonation are attributed to the
344 respective surface morphologies and distributions of carbonation products produced (21). The

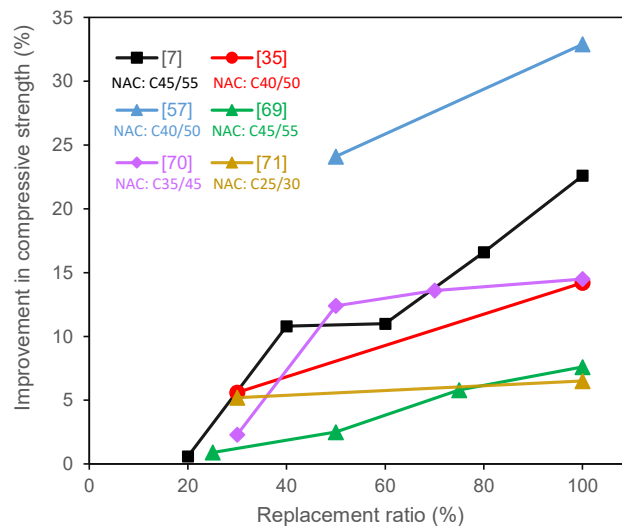
345 respective microstructural changes would influence the chemical interaction between the cRCA
 346 and the new mortar, ultimately affecting the micromechanical properties of the new ITZ. Studies
 347 have shown that carbonation products formed during pressurized moist carbonation are
 348 distributed in layers (66-69). Unreacted hydration products are surrounded by silica gel, while
 349 calcium carbonate agglomerates and fills the pores to form a dense microstructure (21). In
 350 contrast, during aqueous carbonation, precipitated calcite can react with CO₂ and form Ca(HCO₃)₂
 351 when the solution becomes saturated with CO₂. This would alter the calcium carbonate
 352 precipitation-dissolution balance, leading to a spaced distribution of calcium carbonate
 353 precipitates on the cRCA surface. This texture improves the compatibility between cRCA and the
 354 new cement paste. Aqueous carbonation would therefore favour the formation of more reactive
 355 carbonation products than moist carbonation (21), yielding better improvement in the new ITZ.
 356 However, more research is needed to clarify the reasons for the different carbonate precipitation
 357 with aqueous carbonation, considering the fact that a fraction of carbonates may also precipitate
 358 in the solution instead of the pore structure.

359

360 4.2. Effect of RCA carbonation treatment on concrete performance

361 4.2.1. Mechanical performance

362 The carbonation treatment of the RCA improves the compressive strength of recycled aggregate
 363 concrete (7, 35, 57, 69-71)(Figure 5). Xuan et al. (7) found that carbonation pressure and time had less
 364 effect on compressive strength when above 0.1 bar and 24 hours. The improvement in compressive
 365 strength increases with RCA content when concrete is made with cRCA compared to concrete made
 366 with uncarbonated RCA. The compressive strength of cRCA concrete is still lower than that of NA
 367 concrete in the reported cases in the literature. Lu et al. (57) found that at 100% content of coarse
 368 cRCA, compressive strength at 90 d was even comparable to that of NA concrete. This improvement
 369 in compressive strength allows for a higher proportion of cRCA to be used in new concrete.



370

371 Figure 5. Improvement percentage of compressive strength of concrete (28 days curing) with
 372 different replacement ratio of cRCA compared to concrete made with uncarbonated RCA. The
 373 legend indicates the citations for the data sources and the strength class of the respective natural
 374 aggregate concrete used as reference (i.e., mix with 0% replacement ratio).

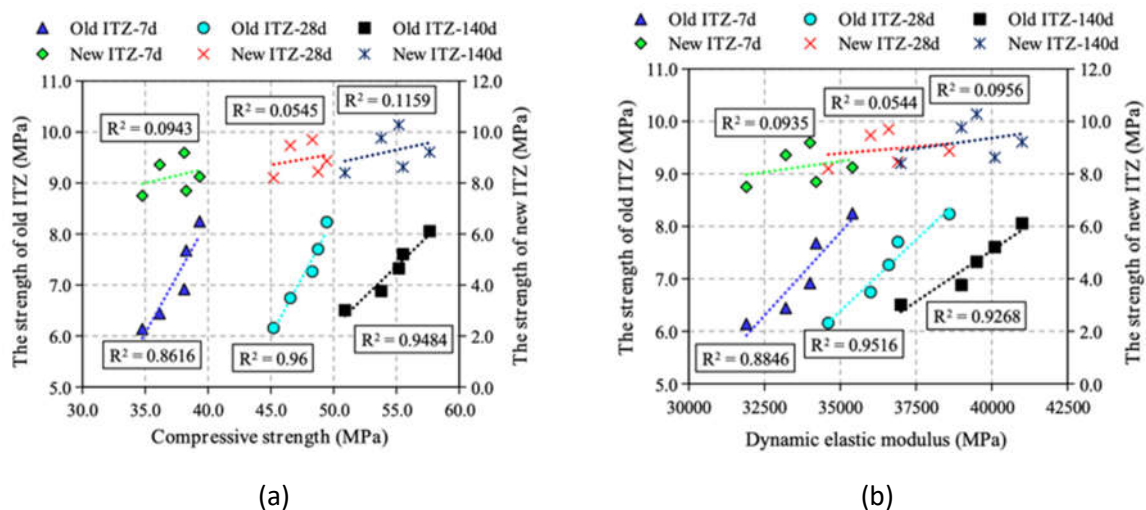
375 The effect of carbonation treatment of RCA on the tensile strength of concrete is generally also
 376 positive, but not as significant as for the compressive strength. For example, Tam et al. (71) found that

377 concrete with 30% cRCA had a higher tensile strength than that of concrete with non-carbonated RCA.
 378 The reduction in porosity and increase in aggregate density are the main drivers for the improvement
 379 with cRCA. These enhancements improve compressive strength more than tensile strength because
 380 density and low porosity are more relevant for the former.

381 Flexural and tensile strength share a similar dependence on bonding characteristics. Most studies
 382 show that cRCA also improves the flexural strength of concrete compared to non-carbonated RCA
 383 concrete (44, 71). Xuan et al. (44) show that 100% cRCA concrete had 28.7% higher flexural strength
 384 than concrete with 100% content of non-carbonated RCA, and 60% cRCA concrete had comparable
 385 flexural strength to that of NA concrete. However, Tam et al. (71) reported a very low absolute flexural
 386 strength (only 2.52 MPa), for the 100% RCA concrete used as a reference, which makes relative
 387 improvements less significant in practical terms.

388 The carbonation treatment of RCA affects the elastic modulus of concrete similarly to compressive
 389 strength. The literature reports 5-29% increases for 100% replacement ratios (7, 35). Being a
 390 volumetric property, the magnitude of the effect on the elastic modulus is connected to the
 391 replacement ratio. Lu et al. (35) found that carbonation treatment of RCA increases the elastic
 392 modulus by 23.2% at a replacement rate of 30% and 28.9% at a replacement rate of 100%, always
 393 relative to uncarbonated RCA concrete. The improvement in elastic modulus was greater than in
 394 density, and this additional positive effect could be interpreted as an improvement in the ITZ. The
 395 improvement was also linked to the carbonation pressure and time. Xuan et. al. (7) found that
 396 increasing the carbonation pressure from 0.1 to 5.0 bar increased the elastic modulus of the concrete
 397 by only 2.3%. Furthermore, when the RAs were carbonated at 5.0 bar for 24 hours, the elastic modulus
 398 of concrete increased by 8.3% compared to concrete with uncarbonated RCA. This is contrary to the
 399 findings concerning effects on compressive strength, and the contrast could be associated with the
 400 improvement of the stiffness of RCA after carbonation.

401 Statistical analysis reveals a strong linear correlation between the carbonation strengthening of the
 402 old ITZ and the compressive strength and dynamic elastic modulus of concrete (64) (Figure 6, a, and
 403 Figure 6, b, respectively). The positive impact on the microhardness of old ITZ (20) is higher for
 404 aqueous carbonation in calcium hydroxide solution than with moist carbonation (24, 64, 72).



405
 406
 407

Figure 6. Relationship between strength of ITZ and mechanical properties of concrete (64).

408 **4.2.2. Durability performance**

409 The durability of concrete prepared with cRCA is associated with changes in the microstructure of the
410 old adhered mortar and the ITZ in cRCA. Commonly accepted durability indicators include drying
411 shrinkage, transport coefficients, carbonation depth, chloride diffusion coefficient, freeze-thaw
412 resistance, and corrosion rate. However, water permeability, gas permeability, water vapor, and gas
413 diffusion coefficient were rarely measured in the literature for concrete with cRCA. Figure 7 presents
414 variations reported in the literature for durability indicators for cRCA concrete compared to non-
415 carbonated RCA concrete (100% replacement). It is important to note that this comparison does
416 not differentiate data based on carbonation duration, CO₂ concentration, external relative humidity, or
417 material age.

418 With the RCA carbonation treatment, most durability indicators show improved values. For drying
419 shrinkage, water absorption, sorptivity, electrical conductivity, and carbonation depth, average
420 reduction rates lie between 10 and 20%. Regarding the freeze-thaw resistance, studies generally
421 reported a reduction in the mass loss, but both increased and reduced dynamic modulus values are
422 found in the literature (73, 74).

423 Drying shrinkage is generally measured by maintaining the young-age prisms at constant relative
424 humidity (50% (44), 60% (39, 64) or 70% (57)) and monitoring length changes over time. Reduction in
425 drying shrinkage with cRCA shows the most notable changes occurring in the first weeks of the test,
426 and then the relative improvement reduces over time (39). In theory, different ranges for
427 improvement would be expected for the various RH setups. However, experimental results from the
428 various studies do not show a clear trend in this sense. The CO₂ concentration during the carbonation
429 treatment affects the drying shrinkage: a higher CO₂ concentration yields a greater reduction in
430 shrinkage. For instance, the reduction in drying shrinkage for samples carbonated at above 99% CO₂
431 concentration is between 23% and 25.2% at the age of 112 days (44), while the reduction is only 11.5%
432 for samples carbonated at 20% CO₂ concentration at the same age (64). Compared with natural
433 aggregate concrete, contradictory results (both reduction and increase) for drying shrinkage of cRCA
434 concrete are reported in the literature. For instance, a decrease of 31% was reported by (75), but an
435 increase of up to 30% was found in other studies (57). The differences may be attributed to various
436 factors, including the quality and the initial condition of cRCA, specimen sizes, and replacement ratios.

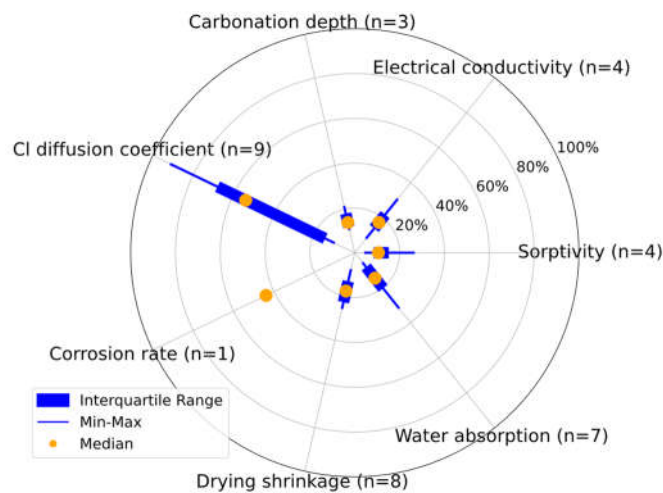
437 Reductions in porosity are revealed by water absorption data (Figure 7). Water absorption is generally
438 measured by fully immersing the concrete sample in water. The average water absorption reduction
439 with cRCA is about 13.5%. Sorptivity measurement is also commonly used, as it is a straightforward
440 method to gauge the durability performance of concrete. Only the first stage of the sorptivity process
441 is considered, disregarding the measurements for the second phase that are influenced by factors
442 other than solely capillary action (76, 77). As shown in Figure 7, carbonating the RCA reduces the initial
443 sorptivity of concrete by about 13% (49, 78-80). This decrease in sorptivity suggests that concrete with
444 cRCA offers greater resistance against water penetration than non-carbonated RCA concrete. The
445 reduction rate in sorptivity coefficients correlates with carbonation duration, with a 7-d treatment
446 showing three times the reduction of a 1-d treatment for concrete carbonated at 100% CO₂ (49).

447 Reductions in transport properties are also reported for other durability indicators such as electrical
448 conductivity and chloride diffusion coefficient. The observed electrical conductivity decreased
449 between 10% and 30%, with an average of 18% (44, 49, 79), aligning well with the sorptivity results.
450 The chloride diffusion coefficient was found to be much more reduced than other transport
451 properties, averaging 47% reduction (49, 64, 78, 79, 81), with some studies reporting a drop of even
452 80-90% (39). The causes of such marked differences are not totally clear, but one of them might be
453 the densification caused by calcium carbonate precipitation in attached old mortar after carbonation

454 treatment (78). The measured corrosion rate induced by chloride ingress is seen to be reduced to a
455 similar extent, about 44%, as shown in Figure 7, since fewer chloride ions can reach and remain at the
456 steel-concrete interface (81). With a reduction in accessible porosity, CO₂ diffusion also slows down,
457 leading to a slower carbonation rate in cRCA compared with non-carbonated RCA concrete (79, 82).

458 Due to their higher alkali content, the use of RCA in concrete may pose a risk in terms of alkali silica
459 reaction susceptibility. In this respect, carbonation treatments may mitigate the risk of ASR by
460 neutralizing the RCA alkalinity with CO₂ and reducing the permeability of the RCA. A recent study
461 reported that RCA from concrete with reactive natural aggregates that was subjected to carbonation
462 showed significant reductions (20-50%) in measured expansions, possibly caused by ASR (83). As such,
463 carbonation treatment, in combination with a low-alkali binder or blended cement, holds promise for
464 recycling ASR-affected RCA in new concrete (84).

465 Overall, all the findings in the literature regarding durability indicators reveal that cRCA offers a better
466 contribution to the durability performance of concrete compared to non-carbonated RCA. The
467 reduction of the accessible porosity and densification of the old ITZ are the main influencing factors.



468

469 Figure 7. Relative reductions in values for concrete durability indicators due to carbonation
470 treatment of RCA, compared with mixes containing non-carbonated RA. The higher the value, the
471 more likely the improvement in concrete durability due to the carbonation treatment. Data collected
472 from (39, 44, 49, 57, 64, 78-82).

473 5. Carbonated RCA in uses other than concrete

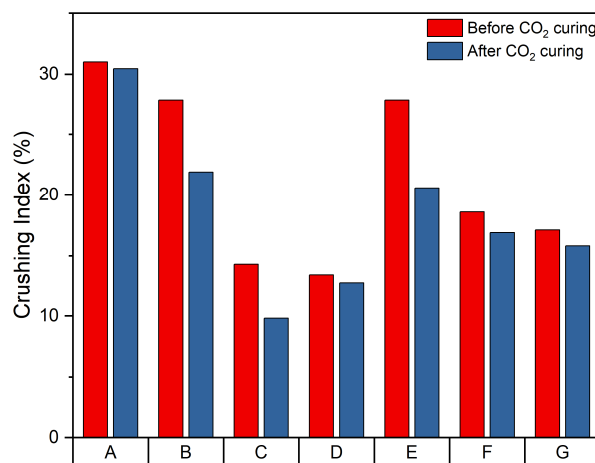
474 The property improvements of cRCA make them attractive for applications other than concrete, for
475 example in screeds, mortars, asphalt concrete, and geotechnical applications such as soil stabilisation
476 and backfilling. Whilst RCA is increasingly being chosen as a replacement for NA in these applications,
477 there remains little research on the use of cRCA. As with concrete, the drawbacks of RCA in these
478 applications are often linked to poor ITZ properties, porosity, high water absorption, and low strength.
479 However, carbonation treatment could lead to superior performance.

480 For example, the fragile ITZ and high porosity of RCA mean that screeds that use RCA achieve a lower
481 strength than those produced with NA (85). Carbonated RCA, with its superior ITZ properties and
482 lower porosity, could be a viable alternative. A lower porosity would reduce the amount of pre-wetting
483 water required; however, the pre-wetting condition of high porosity RCA means that screeds require
484 less additive (85). Pre-wet RCAs have an adhered water film, which prevents flocculation of fines and

485 helps maintain consistency (85). This benefit may be reduced if lower porosity cRCA is used, but this
486 could be outweighed by the potential strength improvements provided by cRCA. Using cRCA may not
487 reach the strengths achieved with NA, but it is likely to produce materials with a higher strength than
488 non-carbonated RCA.

489 Good quality fine aggregates such as river sand are in short supply worldwide (86), so it has become
490 important to identify alternative fine aggregates for use in applications such as mortars. Mortars
491 produced using cRCA have shown improved workability and less shrinkage in comparison to those
492 produced with non-carbonated RCA (39, 87) and can achieve similar flowability and compressive
493 strength to natural sand mortar (39). As for concrete, the improvements can be attributed to the
494 improved quality for the attached mortar and the old and new ITZs (39). Considering the difficulties in
495 accurately determining the water absorption capacity of recycled concrete fines (88, 89), the
496 reduction in porosity upon carbonation treatment is consistent with less variability in the consistency
497 of mortar.

498 The use of RCA in bituminous mixes has been identified as a sustainable approach for producing
499 pavements (90, 91). However, they provide inferior performance due to the higher water absorption,
500 higher crushing characteristics, and interfacial zones associated with the cement mortar adhered to
501 the aggregate surface (91, 92). As carbonation of RCA's reduces water absorption, improves ITZ
502 properties, and decreases the crushing value (41), cRCA is likely to perform better in asphalt than RCA.
503 However, to date there have been no studies performed to compare their performance.



504

505 Figure 8. Crushing values of various RCA types before and after carbonation treatment in different
506 studies. Data collated from (87).

507 While coarse RCA received the most attention for application in concrete, in current practice recycled
508 concrete fines are usually downcycled in geotechnics and backfilling (93-95). In these unbound
509 applications, the crushing value of aggregates is an important property to consider (96). This
510 represents the aggregate's relative resistance to crushing under a gradually increasing compressive
511 load, with a smaller value indicating that a material can exert a stronger resistance under a certain
512 load (96). The crushing behaviour of RCA differs in comparison to other materials due to the adherent
513 mortar, which appears to be crushed at inter-particle points under loading. This behaviour gives a low
514 initial stiffness but eventually allows better contact between stiff core aggregates to develop (97). As
515 shown in Figure 8, carbonation curing can be seen to consistently lower the crushing values of various
516 types of RCA, although the extent of the improvement varies. So, when using cRCA for backfilling and
517 other unbound applications, the initial stiffness should be higher as the mortar will be harder than in

518 non-carbonated RCA. Work investigating the use of RCA as backfilling materials has also identified that
519 RCA that is well compacted at an optimum water content can achieve stiffness as good as well graded
520 gravelly soils (97). Under these conditions, moulding water content and saturation have little effect
521 on strength and deformation characteristics (97). A key aspect to consider is related to the added
522 value for geotechnical applications. Backfilling is normally considered downcycling for RCA; thus, for
523 cRCA, the degree of downcycling will be even higher. The main advantage of cRCA in geotechnical
524 applications seems to be the possibility of storing CO₂ in them, as enforced carbonation helps to bind
525 additional amounts of CO₂ compared to those under natural carbonation.

526 The chemical stabilisation of natural soils with unsatisfactory engineering properties is another
527 potential application for cRCA. RCA can be used to chemically stabilise soils through reactions that
528 modify soil properties (98). When RCA is used to stabilise clay, ion exchange between the RCA and
529 clay produces large aggregates through agglomeration and weak bonding at particle contacts (99).
530 After 1 day, C-S-H and C-A-H form and in the long term more C-S-H and C-A-H forms via reaction
531 between the clay's silica and alumina and the RCA Ca²⁺ (99). RCA can be used alongside other additives
532 such as lime (98) and fibres (99) to improve stabilisation. The addition of RCA can improve stiffness
533 and strength and decrease shrinkage in stabilised clays (100). The improvement in stiffness is
534 attributed to better soil-aggregate interfaces when RCA is used, and superior shrinkage performance
535 is attributed to the high porosity (100). Carbonation treatment reduces porosity and ionic strength, so
536 if cRCA is to be used, it will be necessary to examine how this would impact the properties of stabilised
537 soils. It would also be important to establish the influence that carbonation would have on the
538 reactions that occur. Whilst carbonation-cured concrete can undergo post-carbonation hydration
539 reactions to create a high-strength product with enhanced pore structure, this is usually achieved by
540 fully submerging the carbonated products in water (101). Consequently, it is important to evaluate
541 whether sufficient water for hydration would be or could be made available in the soil stabilisation
542 process and whether the extent of the reactions would be sufficient to produce the desired properties.

543 6. Upscaling and cost/benefit analysis/LCA (capabilities, needs)

544 6.1. Industrial production and application of cRCA

545 An industrial setting for producing cRCA should ideally integrate the following three components with
546 distinct functions:

547 6.1.1. Production and treatment of RCA

548 There are mainly two approaches for selecting the type of RCA source for carbonation. The input
549 materials for carbonation can be either recycled concrete fines or coarse RCA (10, 102). Existing
550 technologies that work with recycled concrete fines and coarse RCA differ in terms of their industrial
551 implementation and the final product. Recycled concrete fines, due to their finer particle size
552 distribution, can be converted to use as a filter bed or slurry tank for treating industrial flue gases
553 containing CO₂. Carbonated recycled concrete fines produced as a result of this can be later used as a
554 supplementary cementitious material in concrete production (103). The second approach, with
555 coarser RCA, tends to carbonate the aggregates in a carbonation chamber so that they can be used as
556 an aggregate for concrete production. There is also another approach where crushed concrete waste
557 is treated in the presence of CO₂ to separate coarse and fine particles. The coarser particles can be
558 used as the aggregates, and the finer particles can be used as carbonated filler or supplementary
559 cementitious material depending on the reactivity.

560 6.1.2. Carbonation of RCA

561 The carbonation chamber essentially has the purpose of exposing RCA to carbon dioxide in the most
562 efficient manner. Apart from the type of RCA source and capacity, operational parameters such as

563 relative humidity, temperature, type of carbon dioxide source, concentration, and time of exposure
 564 need to be considered while designing the carbonation chamber. The carbonation chamber designs
 565 have evolved from closed stationary containers (104) to rotating drum-type and fluidized bed reactors
 566 to improve the efficiency of carbonation (8, 105). The comparison with laboratory results shows
 567 equivalent carbonation at the industrial scale (8, 104, 105). The feasibility of the direct reuse of carbon
 568 dioxide from the flue gases into carbonation chambers will depend on the carbon dioxide
 569 concentration and the exact composition of the flue gas (30). Moreover, the industrial-scale
 570 carbonation chambers need to be compliant with the health and safety requirements of the industrial
 571 operational environment.

572 **6.1.3. Application of cRCA in concrete**

573 The industrial application of cRCA in cement concrete is mainly governed by factors like national and
 574 regional regulations on the raw materials for concrete, the cost of producing cRCA, and the technical
 575 and environmental performance of concrete mixtures with cRCA. National and regional standards
 576 have widely differing approaches toward the use of RCA in concrete. Currently, in many locations, the
 577 cost of production of RCA is higher than that of NA, and this affects the commercial viability of such
 578 aggregates (106). Policy changes promoting the use of cRCA would help to overcome some of these
 579 challenges. There is also a need for studying the long-term performance and durability of concrete
 580 with carbonated recycled concrete fines. Such studies would provide more clarity on selecting the
 581 target applications of concrete containing cRCA following a performance-based approach. Generally,
 582 the use of cRCA instead of non-carbonated RCA improves concrete performance (41, 57, 105, 107).
 583 Therefore, at a minimum, cRCA can be used as RCA.

584 **6.2. Reduction in global warming potential (GWP) associated with cRCA**

585 There are only a few LCA studies or calculations on GWP reductions for producing cRCA. Available data
 586 for GWP reduction the carbonation processes given in the literature are summarised in Table 2. The
 587 CO₂ balance must include emissions during the processing (crushing, carbonation), transport, as well
 588 as avoided impacts and stored CO₂. Generally, the carbonation processes emit between 1 and 5 kg
 589 CO₂-eq and uptake between 7 and 40 kg CO₂ per tonne of cRCA produced. If the process is optimized,
 590 e.g., allowing for more aeration in the rotating drums, there are more emissions from the carbonation
 591 process, but also leads to higher CO₂ uptake, and the CO₂-eq is further reduced about 30 kg per tonne
 592 of cRCA compared to the stationary containers. This only accounts for measurable CO₂, a full
 593 assessment over the life cycle should extend even further as long as cRCA-containing products may
 594 still absorb CO₂ through carbonation during their use and end-of-life phases.

595 Table 2. Summary of the CO₂ uptake, CO₂ eq of the carbonation process and liquefaction, and the CO₂
 596 eq for the production of 1 tonne of cRCA

	kg CO ₂ emitted from the carbonation process(*)	kg CO ₂ uptake	References	kg CO ₂ -eq net reduction for the 1 tonne of cRCA (not including avoided impacts or transport)	kg CO ₂ -eq net reduction for the 1 tonne of cRCA (including avoided impacts for waste disposal - not including transport)
Closed stationary container	1.0	7.2	(104)	6.0	10.0
Rotating drums-type and fluidized bed reactors	2.9	39.0	(105)	36.0	40.0

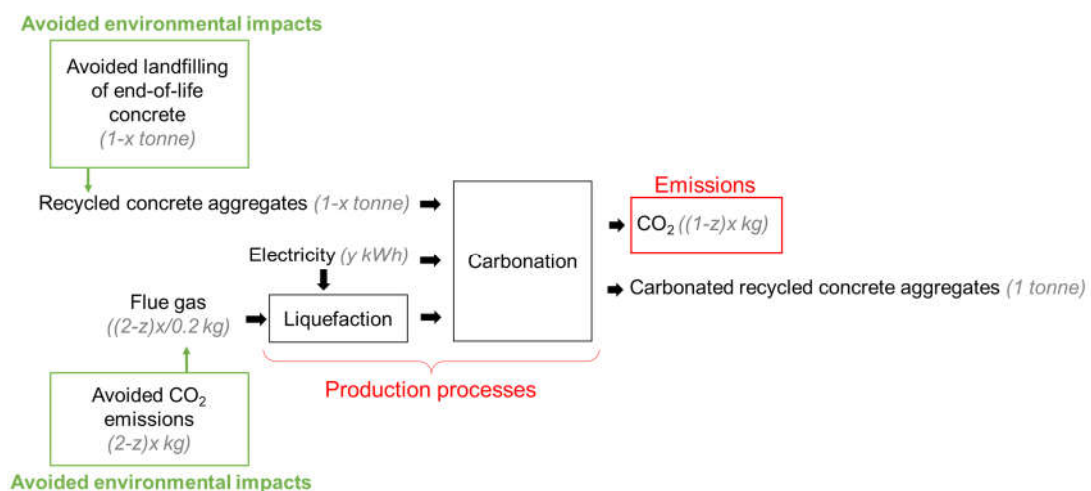
597 (*) calculated from the references (including liquefaction and electricity translated to kg CO₂) or given directly in literature.

598 It would be also convenient to have a separate reporting for CO₂ flows (uptake/release) and avoided
 599 impacts to avoid double-counting of CO₂ savings (e.g., accounting of avoided impact for carbonated
 600 products by the emitting industrial plant with a carbon capture system and the user). A more detailed
 601 reporting would bring more clarity and flexibility for impact allocation decisions. An notable potential
 602 avoided impact of the carbonation upgrading of RCA is related to the improvement in mechanical
 603 performance when compared to concrete containing non-carbonated RCA. This enables for a further
 604 reduction in cement content in concrete while maintaining equivalent performance. Therefore, extra
 605 avoided impact (i.e., less CO₂ emissions associated with the use of cement) are possible when
 606 implemented in concrete mixes.

607 **6.2.1. Consideration of negative CO₂ emissions and avoided impacts**

608 While the CO₂ uptake is evident and can be subtracted from the CO₂ emitted to the air, the CO₂
 609 emissions avoided by treating and recycling the end-of-life concrete (instead of sending it to
 610 landfilling) should be accounted for and specifically reported for the CO₂-eq of a product. The CO₂-eq
 611 can be calculated from the simplified flowchart given in Figure 9, where x kg is the CO₂-eq uptake by
 612 the cRCA, z is the efficiency of CO₂ uptake during the carbonation process, and y is the electricity
 613 required for the CO₂ liquefaction (when storing and transporting the captured CO₂ is also included in
 614 the process) and the carbonation process. The specific values for the GWP depend on the geographical
 615 setting of the processing plant. Finally, this flowchart presents the potential environmental avoided
 616 impacts of this mineral carbonation and reuse of the aggregates, but the GHG emissions related to
 617 transportation are not included here. From the data given in the literature for the CO₂ uptake, the CO₂
 618 related to the carbonation process (considering z=1) and taking into account the avoided impact for
 619 waste disposal, between 10 and 40 kg of CO₂-eq could be saved per tonne of cRCA produced.

620 On top of these numbers, the cRCA can substitute other materials, thereby avoiding their production.
 621 However, accounting for the avoided impacts due to this substitution requires considering the
 622 functional equivalence between the cRCA and the substituted material. It can be considered if primary
 623 aggregates are replaced by cRCA. Most likely, cRCA will replace RCA in concrete. In that specific case,
 624 avoidance of cement addition to reach similar concrete properties can be considered when the use of
 625 cRCA is compared to the use of RCA (108, 109). This depends in turn on the specific applications of the
 626 cRCA. The avoided aggregate production can be added to the calculations; in that case, an extra
 627 reduction of 5 kg of CO₂-eq per tonne of primary aggregate can be considered. This consideration can
 628 enormously change the CO₂-eq of a product, depending on the function of the product replaced.



629

630 Figure 9. Simplified flowchart for cRCA and CO₂-containing flue gas (20% CO₂). Avoided
 631 environmental impacts modeled in this study are indicated in green. x kg is the CO₂-eq uptake by the

632 cRCA, z is the efficiency of CO₂ uptake during the carbonation process, and y is the electricity
633 required for the CO₂ liquefaction (when storing and transporting the captured CO₂ is also included in
634 the process) and the carbonation process. Not displayed: needs in water, wastewater produced, and
635 other chemicals. Transport is not considered in this flowchart. Data for the avoided impacts taken
636 from Ecoinvent v3.8 (110): “treatment of inert waste, inert material landfill” and “market for gravel,
637 round”, Switzerland.

638 *6.2.2. Separation of the fine and coarse fraction in RCA prior to carbonation*

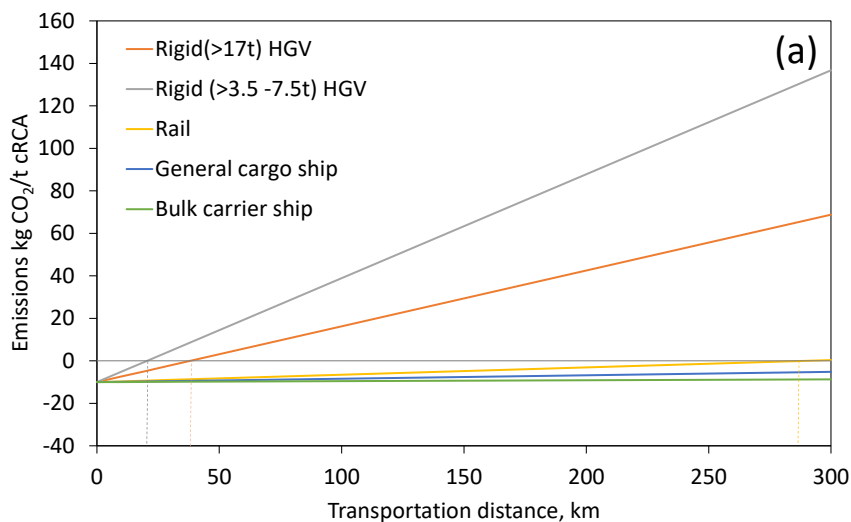
639 Another point is the separation of the fines (rich in cement paste and mortar, hence rich in reactive
640 CaO) from the coarse fraction in the RCA. This can be achieved by the subsequent washing, grinding,
641 drying, and sieving. This process maximizes both the carbonation of the fines and the carbonation of
642 coarse aggregates as it exposes more fresh cement paste. There are then two types of products:
643 carbonated cement paste and coarse cRCA. The carbonated cement paste should be in the near future
644 used as SCMs (103). Such concurrent valorisation increases circularity by using the whole range of
645 particle sizes and increasing the competitiveness of all types of secondary products (111). The quality
646 of the parent concrete has an impact on the efficiency of the process and should be considered.
647 Depending on the strength of the source concrete or the recycling method, more or less cement paste
648 will detach from the coarse fraction (liberation degree). Fines with a concentrated cement paste
649 content serve better to produce alternative carbonated supplementary cementitious materials.

650 The mechanical separation detailed in (112) is estimated at 100 kWh per tonne of cRCA (113),
651 corresponding to 2-10 kg CO₂-eq depending on the source of the electricity. However, the carbonated
652 cement paste also mineralizes a higher amount of CO₂ per tonne of carbonated cement paste. In other
653 scenarios, the functional unit is often 1 tonne of crushed concrete; however, under this specific
654 valorisation strategy, the functional unit product shifts to 1 tonne of detached and carbonated cement
655 paste.

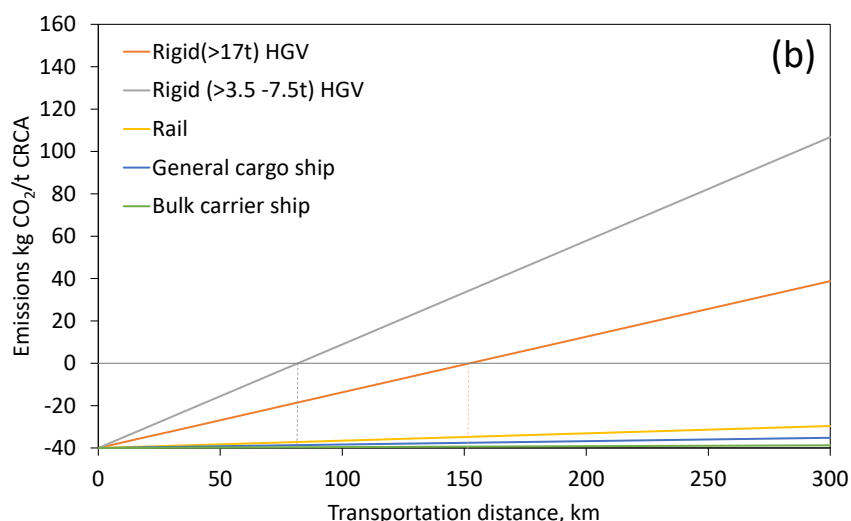
656 *6.2.3. Impact of transport*

657 Aggregates are generally low-carbon products when used locally. Transportation can add considerably
658 to the total CO₂ emitted per unit weight of the material. Considerable variations are reported in CO₂
659 emissions from different modes of transportation. A study conducted by the International Council on
660 Clean Transportation (114) showed that urban delivery trucks with a 4×2 axle configuration emitted
661 on average 0.307 kg CO₂/t-km, which is over five times as much as long-haul tractor-trailers with
662 emissions of 0.057 kg CO₂/t-km. For most vehicle subgroups, the baseline data show a large spread
663 around the mean. For long-haul tractor-trailers (5-LH), there was a 63% difference between the best
664 and worst-performing vehicles. Transportation emissions considered in (105) are selected from the
665 Ecoinvent database, and the value is about the average of the data range in (114). For highly reliable
666 results, studies on transportation GHG emissions specific to the aggregate industry should be
667 conducted and load factors should be tailored to specific scenarios. The use of cRCA involves the
668 transportation of crushed concrete from the recycling site to the carbonation site and also from the
669 carbonation site to the concrete production plant. Figure 10 shows the increase in emissions with the
670 distance of transport per tonne of cRCA for various modes of transportation. Emissions for heavy
671 goods vehicles (HGV), rail, and sea freight are taken from UK government GHG conversion factors 2022
672 (115), with the well-to-tank (WTT) factor added. Fluidised bed and rotating drum technologies allow
673 for longer transportation distances compared to carbonation in closed stationary container, without
674 more GHG emissions than carbonation can uptake. For transportation by ship or rail, reduced GWP
675 (negative values in Figure 10) can be expected even with ~300 km transport distance, especially for
676 processing in rotating drums or fluidised bed reactors. These values are only approximate for this
677 specific scenario, and for each application, evaluation should be conducted on a case-by-case basis.

678 There are net-zero strategies in place for the road freight sector, which depend on innovations in bio-
 679 LNG fuels, battery electric vehicles, hydrogen energy, and low carbon bio-fuels and is on the path of
 680 decarbonization. Given the range of values in the net reduction in GWP per tonne of cRCA, transport
 681 of the RCA and of the subsequent cRCA produced is currently the main limitation related to the
 682 technology. The decarbonization of this sector can further improve the viability of use of technology.



683



684

685 Figure 10. Increase of carbon emissions with supplementary transportation distance for cRCA for
 686 various transportation means, and breakeven points for (a) closed stationary container technology,
 687 and (b) rotating drums or fluidised bed reactors. (HGV- Heavy goods vehicle). NOTE: reductions in
 688 GWP would vary depending on the energy sources for the treatment of the cRCA. Data source for
 689 emissions is (115).

690 6.3. Cost-benefit analysis

691 A cost-benefit analysis (CBA) allows for an evaluation of the economic viability of the carbonation
 692 process. This analysis has to be done over a long period (assuming a typical period of 50 or 100 years
 693 for the carbon cycle from the GHG emission during production of constituents for the construction to
 694 the recycling as cRCA) and therefore requires assumptions about long-term interest rates (116). A first
 695 indication can, however, be obtained by comparing the costs of the process with the short-term
 696 induced benefits.

697 For the estimation of the costs, like for the previous CO₂-eq calculations, the process itself and the
698 transport distances involved must be considered. During the FastCarb project (8), an estimation based
699 on the results of the industrial demonstrator has shown that, depending on the transport distance
700 due to the process, the cost of the cRCA is between double and triple that of non-carbonated RCA (8)
701 (113). This extra cost could neither be compensated by a possible gain on the stored CO₂ nor by the
702 improvement of the properties of concrete made with these cRCA. The carbon reduction
703 commitments of the construction industry could be favourable in overcoming this price difference.
704 Low-carbon cement and concrete products are often marketed as premium products by the
705 producers. These premium low-carbon products are always sold at a higher price than conventional
706 products (117-119), creating additional revenue. Globally, there is a shift in procurement policies,
707 leading to more importance for sustainable public procurement. The same type of product from
708 different suppliers is compared by contractors, both in terms of price and carbon dioxide footprint.
709 Even if the cost of the product is high, a lower carbon footprint will make it a preferred choice among
710 other competing options, due to carbon reduction targets stipulated by clients during the tender
711 stage. In the Netherlands, a ranking method combining these two aspects into an environmental cost
712 indicator is practiced (120).

713 Moreover, the CO₂ stored in these aggregates during the process can be valorised (121). Two ways are
714 possible. The first one is linked to the cement process and a trading system (ETS in Europe). The second
715 one uses the concept of a CO₂ reduction certificate.

716 *6.3.1. Valorisation through a cap and trade system (example: European ETS)*

717 The European Union Emissions Trading System (EU ETS) is a 'cap and trade' system, creating
718 "allowances which are essentially rights to emit GHG emissions equivalent to the global warming
719 potential of 1 tonne of CO₂ equivalent (tCO_{2e}). The level of the cap determines the number of
720 allowances available in the whole system" (122). This system concerns CO₂ emissions from power
721 generators and energy-intensive industries like the cement industry (other sources like the production
722 of bio-methane are not concerned). Free allowances are attributed to each concerned industrial site,
723 but the level of these allowances is decreasing each year and generally does not cover the real
724 emissions. The consequence is that the sites must buy allowances on the market. The result is that the
725 price of CO₂ has risen sharply: from 12€ in March 2018 to 90€ five years later. Considering the amount
726 of CO₂ that could be stored in RCA, it is very interesting for the concrete industry to use carbonated
727 sand (8) and recycled concrete fines (103) to reduce the level of CO₂ they have to buy on the market.
728 In Europe, the standard EN 197-6 "cement with recycled building materials" allows the introduction
729 of composite cements with recycled concrete fines. If these fines are carbonated, the stored CO₂ could
730 be deduced from the emissions of the cement plant, thus valorising the carbonation.

731 *6.3.2. Valorisation through CO₂ reduction certificates*

732 Another way to valorise carbonation, particularly interesting for bio-sourced CO₂, is through the issue
733 of carbon removal certificates. Companies that want to reduce their CO₂ impact but do not have the
734 means to do so or for which it is too expensive can buy a certificate proving that CO₂ has been stored
735 elsewhere. The stored CO₂ has to be quantified through a known methodology validated by an
736 independent organization that can certify the process and how the stored CO₂ is estimated. This
737 market is currently operated by private organisations (some of them being non-profit), but there are
738 reflections for a more normative vision. For instance, the European Parliament has produced a pre-
739 legislative synthesis about the certification of carbon removals that would organize and facilitate the
740 market in the future (123).

741

742 7. Conclusions and perspectives

743 The following conclusions summarize the key findings regarding the potential and bottlenecks for the
744 enforced carbonation of recycled concrete aggregates (RCA):

- 745 • *Motivation and process optimization:* Originally, performance improvement was the driver for the
746 enforced carbonation of RCA. Interest is nowadays partially shifting to CO₂ uptake capabilities.
747 Optimized enforced carbonation, whether for maximum enhancement or maximum CO₂ uptake,
748 can be achieved with specific values for temperature, RCA moisture content and (sometimes)
749 particle size, CO₂ concentration, gas pressure, and duration of exposure. These optimal
750 parameters relate to the type and quality of RCA and the carbonation method used.
- 751 • *Enforced carbonation methods:* Prevalent enforced carbonation methods for RCA include flow
752 through moist (or semi-dry) carbonation, pressurized moist carbonation, aqueous carbonation,
753 and moist carbonation in a rotating drum. Specific features of each method may show advantages
754 towards specific objectives. Aqueous carbonation enables CO₂ uptake the most. As such, it can
755 advantage gas-solid methods for treating fine RCA with a high carbonatable phase content.
756 Another important contrasting feature is the treatment duration, which ranges from as little as 6
757 hours for aqueous carbonation to more than 24 hours for moist methods. Among moist
758 carbonation methods, the flow through method can make efficient use of CO₂ concentrations of
759 up to 20%, while gases with a richer CO₂ concentration would only result in greater CO₂ uptake
760 when applied under high pressure. Moist carbonation enhancement of RCA is also facilitated by
761 moisture (pre)conditioning, continuous contact, and mixing (for example, in rotating drums).
- 762 • *Improvements on the properties of cRCA and cRCA concrete:* Overall, the literature shows that
763 enforced carbonation leads to favourable changes in a wide range of RCA properties (e.g., lower
764 porosity, lower crushing indexes). Nevertheless, there is a need to be careful while interpreting
765 the data, as the observed improvements are not uniform across all conditions and properties. At
766 the concrete level, the degree to which cRCA translates into improved performance of RCA
767 concrete also varies. The primary changes in cRCA that improve concrete performance are the
768 reduced porosity of the old, attached mortar, improvements in the old ITZ in RCA, and
769 improvements in the new ITZ formed with the new matrix. Aqueous carbonation seems more
770 effective than moist carbonation to improve the ITZ, but more research is still needed to clarify
771 the link between precipitated carbonates and ITZ properties with the different carbonation
772 methods. It should be noticed that most of the research has been conducted with coarse RCA
773 fractions, and it is not totally clear if the outcome would be similar for fine fractions of RCA (with
774 greater contents of carbonatable phases). Mechanical performance and transport properties are
775 improved for cRCA concrete in comparison with RCA concrete. Some concerns persist in terms of
776 durability performance. The effects of enforced carbonation on the alkali-silica reaction of reactive
777 RCA, resistance against sulfate attack and acid attack, as well as freeze-thaw resistance have not
778 received sufficient attention. This is relevant not only for applications as aggregate in new
779 concrete, but also for unbound uses of cRCA. Effects on concrete slump and dimensional stability
780 are also areas that require further research. Overall, the properties of cRCA are intermediate
781 between those of the original RCA and reference natural aggregates.
- 782 • *Uses of cRCA in applications different from concrete production:* There is still relatively limited
783 research on the use of cRCA in applications other than concrete. The microstructural
784 improvements should make it more suitable than RCA for application in screed, mortar, asphalt
785 concrete, soil stabilization, and backfilling materials. Literature on all these applications is still
786 scarce. Low-value applications (e.g., soil stabilisation, backfilling) may not require improved RCA

787 properties per se. Still, regardless of the specific application, enforced carbonation may remain
788 interesting in terms of exploiting the CO₂ uptake capacity of RCA.

- 789 • *Environmental impact assessment*: The net benefit in terms of CO₂ uptake requires careful tuning
790 of inventories that give origin to reliable Life Cycle Assessment (LCA). Transportation has a
791 significant impact. For a given type of production technology, emissions of transport beyond
792 certain distances or by certain means may well invalidate CO₂ net savings. Thus, transportation
793 deserves remarkable attention from research towards upscaling.
- 794 • *Upscaling challenges*: Upscaling the carbonation process to meet the demand for aggregates from
795 large-scale construction projects is a pending subject. Most of the current studies still focus on
796 laboratory-scale experiments, with limited research on the feasibility and effectiveness of
797 enforced carbonation in industrial settings. The critical aspects of the context, such as local
798 regulations, market conditions (including the availability of local resources for aggregates), and
799 production costs, must be addressed. Primary data is vital to corroborate that the implementation
800 of cRCA is a cost-effective and sustainable solution for construction projects. Recent cost-benefit
801 analyses show that implementing cRCA can duplicate or triplicate costs with respect to
802 conventional RCA. With these high costs, strategies for increasing product competitiveness are
803 needed (e.g., market value of premium low-carbon products, compliance with stricter sustainable
804 public policies, and capitalisation of stored CO₂ through trading systems or CO₂ reduction
805 certificates). Without monetizing such strategies, it seems quite difficult that cRCA can be brought
806 to massive application in new construction in the near future.

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