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1 **Effect of pre-conditioning on carbonated reactive MgO-based concrete samples**

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12
13 **Abstract:** This study investigated the influence of pre-conditioning on the strength and
14 microstructural development of carbonated reactive MgO cement (RMC)-based concrete mixes.
15 The hydration mechanisms of the prepared formulations were studied via isothermal calorimetry.
16 Compressive strength and porosity measurements were conducted to assess sample
17 performance. X-ray diffraction (XRD), thermogravimetric analysis (TGA) and scanning electron
18 microscopy (SEM) were used for the microstructural analysis and quantifications of phases within
19 each sample. Subjecting RMC samples to moderately elevated temperatures (50-60°C) for 1-2
20 days before the start of the curing process enhanced the hydration process. This increase in the
21 degree and rate of the hydration reaction increased the amount of phases available for the
22 subsequent carbonation reaction. The increase in the content and size of hydromagnesite
23 ($4\text{MgCO}_3 \cdot \text{Mg}(\text{OH})_2 \cdot 4\text{H}_2\text{O}$) crystals led to denser microstructures, thereby facilitating higher
24 strengths in samples subjected to pre-conditioning. The obtained results showed that the
25 application of this practical approach to the preparation of RMC-based samples can not only
26 present a more efficient use of the binder component, but also enable the increased
27 sequestration of CO_2 within these samples.

28
29
30 **Keywords:** Reactive MgO cement; pre-conditioning; carbonation; strength; microstructure

31 1. Introduction

32

33 The high environmental impact of the production of Portland cement (PC) necessitates the
34 development of more sustainable solutions. One of the initiatives include the use of reactive MgO
35 cement (RMC), which can offer certain sustainability and performance advantages, depending on
36 the production and curing routes used (Aïtcin 2000, Li 2013, Unluer and Al-Tabbaa, 2014).

37

38 The hydration product of RMC is brucite (i.e. magnesium hydroxide, $\text{Mg}(\text{OH})_2$), which is not a
39 major strength resource for RMC-based mixes (Vandeperre et al. 2008). However, when
40 carbonated, RMC-based mixes were reported to gain significant strength (Liska and Al-Tabbaa
41 2009, Liska 2010, Mo et al. 2017, Yi et al. 2013, Dung and Unluer 2018, Dung and Unluer 2019).
42 Hydrated RMC can react with CO_2 to form hydrated magnesium carbonates (HMCs) with different
43 properties. The absorbed CO_2 can be permanently stored in RMC, thereby enabling its long-term
44 sequestration (Dung and Unluer 2016, Unluer 2012, Liska 2010, Unluer and Al-Tabbaa 2011).
45 Some of the main HMCs observed in carbonated MgO systems are nesquehonite ($\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$),
46 hydromagnesite ($4\text{MgCO}_3 \cdot \text{Mg}(\text{OH})_2 \cdot 4\text{H}_2\text{O}$), dypingite ($4\text{MgCO}_3 \cdot \text{Mg}(\text{OH})_2 \cdot 5\text{H}_2\text{O}$) and artinite
47 ($\text{MgCO}_3 \cdot \text{Mg}(\text{OH})_2 \cdot 3\text{H}_2\text{O}$). Carbonation is critical for RMC mixes as their strength development
48 mainly depends on the formation of HMCs. The formation of HMCs is correlated with volume
49 expansions, which results in an increase in sample density and a reduction in overall pore volume;
50 and microstructural evolution as the carbonate crystals contribute to the binding strength via the
51 continuous network structure they form.

52

53 One challenge presented by RMC formulations is the low dissolution of MgO under ambient
54 conditions (Amaral et al. 2010, Fruhwirth et al. 1985, Vermilyea 1969), which can limit the
55 subsequent hydration and carbonation reactions. This was shown by previous studies (Unluer
56 and Al-Tabbaa 2014, Thomas et al. 2014, Tang et al. 2014), where the maximum hydration of
57 MgO under ambient conditions was reported as $\sim 80\%$. Furthermore, the precipitation of initial
58 reaction products on the unreacted MgO particles can prevent the continuation of

59 hydration/carbonation (Dung and Unluer 2018, Dung and Unluer 2017c). These limitations result
60 in low strength development and the inefficient use of RMC as a binder.

61

62 The use of moderately elevated temperatures before or as a part of the curing process could
63 present a solution to these limitations. Accordingly, previous studies (Wang et al. 2014, Lin and
64 Meyer 2009) reported an increase in the reaction rates with an increase of 10°C in the reaction
65 temperatures. In PC-based samples, the use of elevated curing temperatures was shown to
66 increase the early-age strength and provide enhanced resistance to aggressive environments via
67 the increased formation of dense hydrate phases (Khatib and Mangat 2002, Lothenbach et al.
68 2007). While elevated curing temperatures can accelerate the initial hydration process, the
69 increase in density via the rapid precipitation of hydration products can hinder the permeation
70 of free water. This effect can slow down the hydration rate in later ages and result in a non-
71 uniform microstructure (Lin and Meyer 2009, Wang et al. 2012).

72

73 Pre-conditioning of cement-based samples is one of the initiatives that involve the use of
74 moderately elevated temperatures ranging from 20°C to 80°C under a relative humidity (RH) of
75 50-98% (Zhan et al. 2012, Morshed and Shao 2013, Shi and Wu 2008). The reduction in the
76 amount of extra water in the pore space via pre-conditioning can increase the permeability of
77 concrete samples, thereby enabling a higher degree of carbonation and faster strength
78 development in shorter curing durations. However, previous studies reported inconsistent
79 outcomes on the use of pre-conditioning on PC-based samples due to the differing role of high
80 temperature and humidity in influencing the reaction mechanisms. Accordingly, while elevated
81 temperatures lead to increased reaction rates, a reduction in the solubility of CO₂ can also be
82 observed (Vorholz et al. 2000), which can lower the dissolution rate of Ca ions in the high pH of
83 the pore solution. Furthermore, a high humidity can enhance the hydration process, whereas the
84 penetration of CO₂ can slow down under saturated environments. Another undesirable outcome
85 is the formation of cracks under the elevated temperatures used during this process, which can
86 be compensated with high humidity. Therefore, a careful consideration of all the parameters
87 used during pre-conditioning is needed to optimize sample performance.

88

89 Although pre-conditioning has been used in accelerating the strength development of PC-based
90 samples, this method has not been applied to RMC-based samples until now. Considering the
91 limitations in the reaction mechanisms of RMC formulations, pre-conditioning can provide the
92 necessary environment to enhance hydration and carbonation reaction kinetics and thereby
93 effectively improve sample performance. Accordingly, this study investigated the influence of
94 pre-conditioning involving the use of different temperatures on the strength and microstructural
95 development of RMC-based concrete samples. The hydration mechanisms of the prepared
96 formulations were studied via isothermal calorimetry, whereas the degree of hydration was
97 quantified via thermogravimetric analysis (TGA). Compressive strength and porosity
98 measurements were conducted to assess sample performance. These results were further
99 supported by a detailed microstructural analysis and phase quantifications via x-ray diffraction
100 (XRD), TGA and scanning electron microscopy (SEM).

101

102

103 **2. Materials and Methodology**

104

105 **2.1. Materials and sample preparation**

106

107 RMC was used as the main cement binder in this study, with a commercial name “calcined
108 magnesite 92/200”, obtained from Richard Baker Harrison Ltd (UK). The chemical composition of
109 RMC is shown in Table 1.

110

111 The water/cement (w/c) ratios of RMC samples were set at 0.6, in line with the standard
112 consistency of RMC. The mix design used in this study included 40% cement and 60% coarse
113 aggregates (i.e. by mass). The coarse aggregates were composed of granite chippings with a
114 particle size ranging between 4.7 mm and 9.5 mm. No fine aggregates were included in the
115 preparation of concrete samples to avoid the presence of silica in the carbonated paste extracted
116 from concrete samples. While the cement content of the prepared samples was higher than that

117 of conventional concrete mixes due to the lack of fine aggregates, this approach was purely taken
118 to increase the accuracy of the quantification of hydrate and carbonate phases performed via
119 XRD and TGA, which helped with the understanding of reaction mechanisms. A similar mix design
120 was adopted in previous studies (Pu and Unluer 2018, Ruan and Unluer 2017b, Dung and Unluer
121 2016). 50x50x50mm concrete samples were cast and consolidated by a vibrating table and trowel
122 finished. After casting, samples were kept in an ambient environment for 24 hours until
123 demolding. Samples subjected to pre-conditioning were placed in an environment chamber,
124 whose temperature was set between 40°C and 60°C at a fixed RH of 90%, for 1 or 2 days. The
125 pre-conditioning process was followed by the curing of samples under a CO₂ concentration of
126 10%, temperature of 30±2°C and RH of 80±5% for up to 56 days. In addition to the concrete
127 samples, a separate batch of paste samples (i.e. no coarse aggregates) was prepared and cured
128 under the same environment as the concrete samples to study the changes in the hydration
129 mechanism under pre-conditioning.

130

131

132 **2.2. Methodology**

133

134 **2.2.1. Isothermal calorimetry**

135

136 The heat flow and cumulative heat associated with the hydration of each mixture were recorded
137 at 30-60°C by an I-Cal 2000 High Precision Calorimeter, according to ASTM C1702-15a (ASTM
138 2015). To initiate the analysis, the prepared pastes were immediately placed into the calorimeter
139 channel to record their heat of hydration for up to 48 hours.

140

141

142 **2.2.2. Compressive strength**

143

144 The compressive strength of RMC concrete samples cured for different durations (i.e. up to 56
145 days) was measured using uni-axial loading, according to ASTM C39/C39M-16 (ASTM 2016). The

146 equipment used was a Axiomtek compressing machine, which was set at a loading rate of 50
147 kN/min.

148

149

150 **2.2.3. Porosity**

151

152 The porosities of the samples at different durations of curing were calculated via Equation 1 in
153 line with ASTM C642-06 (ASTM 2006), where m_{sat} is the sample mass saturated in water, m_{dry} is
154 the sample mass dried at 70 °C for 3 days and v is the sample volume.

155

$$156 \quad p = \frac{m_{sat} - m_{dry}}{v} * 100 \quad (1)$$

157

158

159 **2.2.4. Microstructural analysis**

160

161 Samples for microstructural analysis were extracted from the concrete cubes crushed during
162 compressive strength testing (i.e. the region surrounding the coarse aggregates) and broken
163 down to small fragments with a clean hammer to the specific size required for each
164 microstructural analysis technique (e.g. XRD, TGA and SEM). Coarse aggregates were not included
165 in these selected fragments for an accurate determination of the contents of each phase (e.g.
166 MgO, brucite and HMCs) within each sample. These extracted fragments were stored in acetone
167 for 3 days to stop hydration, followed by vacuum drying for another 7 days. The vacuum dried
168 samples prepared for XRD and TGA were crushed into powder form and sieved through a 125 μ m
169 sieve. The XRD analysis was performed using a Bruker D8 Advance powder diffractometer with
170 Cu K_{α} radiation, operated at 40 kV and 30 mA. The scanning rate was set at 1° 2 θ /step from 10
171 to 80° 2 θ . Phase contents in each sample were measured by Rietveld analysis with TOPAS 5.0
172 software based on fundamental parameter approach. TGA was performed using a Perkin Elmer
173 TGA 4000 equipment. Samples were heated from 40°C to 800°C at a heating rate of 10°C /min
174 under nitrogen flow. The hydration degrees (HD) of the prepared samples were also evaluated

175 by TGA, in line with Equation 2, where L represents the weight loss corresponding to the
176 dehydration of brucite; C represents the MgO content as a percentage of the total cement
177 component; and 40 and 18 are the molecular weights of MgO and H₂O, respectively.

$$178$$
$$179 \text{ HD} = [L \times 40/18]/[1 - 0.01L) \times C] (\%) \quad (2)$$
$$180$$

181 Finally, the morphologies of the hydration and carbonation products within the prepared
182 samples were investigated via SEM carried out with a Zeiss Evo 50 microscope.

183

184

185 **3. Results**

186

187 **3.1. Isothermal calorimetry**

188

189 The heat flow and cumulative heat of RMC pastes over 48 h of hydration are presented in Fig.
190 1(a) and (b), respectively. As shown in Fig. 1(a), heat flow increased in the early stages, indicating
191 that dissolution took place a few minutes after mixing. The dissolution of the control sample
192 (30°C) was associated with the lowest heat flow, whereby increasing the temperature increased
193 the pace of this process. When compared to the control sample, the significantly higher and
194 sharper exothermic peaks of samples subjected to 40-60°C confirmed the enhancement of the
195 hydration of MgO under these moderately elevated temperatures. This trend became more
196 obvious as the temperature increased to 50-60°C.

197

198 In line with the heat flow results, the cumulative heat curves in Fig. 1(b) clearly indicated that
199 elevated temperatures led to higher hydration degrees, whereas the control sample revealed the
200 lowest heat. The amount of total heat released increased with an increase in temperature, which
201 could be explained by the increased dissolution of MgO and subsequent precipitation of hydrate
202 phases. From the calorimetry results, samples subjected to temperatures of 50°C and 60°C

203 revealed the highest initial hydration rates, which could contribute to the performance of these
204 binders.

205

206

207 **3.2. Compressive strength**

208

209 Fig. 2 shows the strength development of RMC-based concrete samples at different durations of
210 curing, including the initial 1-2 days of pre-conditioning, followed by carbonation curing for up to
211 56 days. While all samples continued to develop in strength up until 56 days, strength gain was
212 much faster during the first 28 days of curing.

213

214 Before 7 days of curing, all samples revealed similar strength results ranging between 20 MPa
215 and 24 MPa at 7 days. A more obvious difference in strength was observed as the curing duration
216 increased. When compared to the control sample (30°C), samples subjected to pre-conditioning
217 under 40°C did not reveal an improvement in terms of their strength. However, as the pre-
218 conditioning temperature increased to 50°C and 60°C, a notable increase in strength was
219 observed. Accordingly, those subjected to 50°C for 2 days or 60°C for 1 day revealed similar
220 strengths of ~47 MPa at 56 days. Furthermore, samples subjected to 60°C for 2 days achieved
221 higher 56-day strength (50 MPa).

222

223 As the main strength development of RMC-based samples is due to the carbonation of Mg-based
224 phases (Dung and Unluer 2016, Dung and Unluer 2018, Ruan and Unluer 2017a, Ruan and Unluer
225 2017b, Liska 2010, Liska and Al-Tabbaa 2009, Mo et al. 2017), an increase in the hydrate phases
226 available for carbonation was one of the possible reasons for the higher strengths of samples
227 subjected to pre-conditioning. While the isothermal calorimetry results (Section 3.1)
228 demonstrated that the effect of pre-conditioning in accelerating the dissolution of MgO and
229 increasing the subsequent precipitation of hydrate phases was obvious after a few hours of
230 mixing, this did not translate into higher early strengths. Alternatively, the conversion of these
231 hydrate phases into carbonate phases throughout the curing process, which was the main source

232 of strength, led to ~25% higher strengths in the longer term in those samples subjected to pre-
233 conditioning under 50°C and 60°C. Samples subjected to longer pre-conditioning durations of 2
234 days achieved higher strengths when compared to those subjected to 1-day pre-conditioning.
235 This difference in performance was more obvious at longer curing durations (e.g. 56 days).
236 Overall, the strength results highlighted that a notable increase in the strength of RMC samples
237 (i.e. from 40 MPa to 50 MPa) could be obtained by simply subjecting the prepared samples to
238 moderately elevated temperatures (i.e. 50-60°C) for 1-2 days before carbonation curing.

239

240

241 **3.3. Porosity**

242

243 Fig. 3 shows the porosities of RMC-based concrete samples at different durations of curing.
244 Generally, the porosities of all samples decreased with curing duration, which was related to the
245 continuation of hydration and carbonation. The formed hydrate and carbonate phases filled in
246 the initially available pores, which led to a lower overall porosity and increased strength. The
247 initially sharp decline that occurred during the first 28 days of curing was followed with a more
248 gradual decline in porosity up until 56 days, indicating a decrease in the rate of hydration and
249 carbonation with time.

250

251 Out of all the samples, the control sample (30°C) revealed the most stable porosity without any
252 major changes over 56 days of curing. These constant porosity levels could explain the lower
253 strengths of this sample when compared to others. Alternatively, samples subjected to pre-
254 conditioning demonstrated a continuous reduction in porosity over time. Those subjected to
255 50°C and 60°C had the lowest porosities at all curing durations, which was in line with their
256 highest strengths amongst all samples. The reduction in the porosities of these samples from 4.2%
257 at 7 days to 3.9% at 56 days could be due to the initially increased formation of hydrate phases,
258 and the subsequent conversion of these phases to carbonates over time. Accordingly, the use of
259 moderately elevated temperatures for 1-2 days before curing enhanced the dissolution of MgO
260 and the precipitation of hydrates. This higher degree of hydration increased the carbonation

261 potential of these samples, thereby translating into lower porosities, denser microstructures and
262 higher strengths over time.

263

264

265 **3.4. XRD**

266

267 Fig. 4 shows XRD patterns of all RMC samples after 56 days of curing. The presence of residual
268 MgO (i.e. PDF #004-0829, main peak at $43.5^\circ 2\theta$) indicated incomplete hydration in all the
269 samples investigated. Similarly, the brucite (PDF #083-0114) peaks at 18.7° and $38^\circ 2\theta$ revealed
270 the higher carbonation potential of all samples. Other than MgO and brucite, peaks
271 corresponding to the HMC phase hydromagnesite (PDF #025-0513) were observed at $\sim 13.9^\circ$, 15.5°
272 and $30.8^\circ 2\theta$. Although other phases (e.g. nesquehonite and dypingite) could have also existed,
273 the contents of these phases were very limited and therefore the main peaks corresponding to
274 these phases could be hardly observed in the XRD spectrum. The presence of magnesite was also
275 seen in all samples as an impurity due to the incomplete calcination process.

276

277 The brucite peaks of samples subjected to pre-conditioning demonstrated higher intensities than
278 those of the control sample, which could be explained by the enhancement of the hydration
279 process in the presence of moderately elevated temperatures (i.e. $50-60^\circ\text{C}$) during the first 1-2
280 days of curing. This increase in the hydration degree led to a higher amount of hydrate phases
281 available for carbonation. Accordingly, when compared with the control sample, the use of pre-
282 conditioning led to an increase in the intensity of the hydromagnesite peak, indicating the higher
283 degree of carbonation observed within the pre-conditioned samples.

284

285 Table 2 includes the quantities of major phases within all samples, calculated by Rietveld analysis.
286 One of the most obvious observations was the decline in the MgO (i.e. periclase) content with
287 the use of pre-conditioning under $50-60^\circ\text{C}$. While the control sample (30°C) revealed an MgO
288 content of $\sim 24\%$, subjecting the samples to 1-2 days of hydration under moderately elevated
289 temperatures of $50-60^\circ\text{C}$ reduced this content. The MgO content of samples subjected to pre-

290 conditioning at 60°C for 2 days was as low as ~11%, thereby enabling a higher utilization degree
291 of the main binder. The use of pre-conditioning also led to higher brucite contents, which went
292 up from ~33% (at 30°C) to 38% (at 60°C). Combined with the higher utilization of MgO
293 demonstrated by the lower periclase contents, this was an indication of the enhancement of the
294 hydration process under elevated temperatures, albeit the short duration (i.e. 1-2 days) of pre-
295 conditioning. Furthermore, a systematic increase in hydromagnesite content was observed with
296 an increase in the pre-conditioning temperature or duration from 50°C onwards. The rise in the
297 hydromagnesite content from ~40% (at 30°C) to ~48% (at 60°C) could explain the improved
298 performances of these samples, which revealed higher degrees of carbonation via the increased
299 dissolution of MgO and precipitation of hydrate phases under elevated temperatures.

300

301

302 **3.5. TGA**

303

304 Fig. 5 shows a representative mass loss and heat flow curve of RMC samples after 56 days of
305 curing. The decomposition of hydration and carbonation products in RMC samples was
306 associated with the release of water and CO₂, reflected by the individual endothermic peaks.
307 Based on the mass loss patterns, the decomposition steps of all samples can be divided into 3
308 main stages, in line with previous literature (Vágvölgyi et al. 2008, Hollingbery and Hull 2010,
309 Frost and Palmer 2011, Jauffret et al. 2015, Ballirano et al. 2010, Ballirano et al. 2013, Teir 2009):
310 (i) the loss of unbound water and the water of crystallization of hydromagnesite due to its
311 dehydration at 40-300°C; (ii) the decomposition of uncarbonated brucite and dehydroxylation of
312 hydromagnesite at 300-500°C; and (iii) the decarbonation of hydromagnesite and magnesite at
313 500-800°C.

314

315 While a summary of these different stages of thermal decomposition is provided in Table 3, the
316 overlap between these decomposition reactions must be acknowledged, therefore presenting a
317 challenge in defining fixed temperature ranges for each reaction. Keeping this challenge in mind,
318 Table 4 presents the mass loss values observed in all RMC samples after 56 days of curing. Mass

319 loss within all temperature ranges increased with an increase in the pre-conditioning
320 temperature (i.e. from 30°C to 50-60°C) or duration (i.e. from 1 to 2 days), which was in
321 agreement with the isothermal calorimetry and XRD results. The higher mass loss in samples
322 subjected to pre-conditioning indicated the increased formation of hydration and carbonation
323 products, which was one of the reasons for the higher strengths achieved by these samples.

324
325 The TGA results were used to obtain the hydration degrees of all RMC samples at different
326 durations, as shown in Fig. 6. Accordingly, the control sample revealed a limited hydration degree
327 of 52% over 7 days. Subjecting the samples to pre-conditioning at 40°C revealed an increase in
328 the hydration degree from 23% to 29% at as early as 1 days, which reached a value of 61% at the
329 end of 7 days. However, the biggest difference was seen when the pre-conditioning temperature
330 was increased to 50°C, revealing a hydration degree of 45% after only 1 day. This value increased
331 to 52% when the samples were pre-conditioned under 60°C, eventually reaching an overall
332 hydration of 69% at 7 days. The rate of change of the hydration degree was higher during the
333 first 3 days, followed by a gradual increase afterwards, which was an indication of the immediate
334 effect of pre-conditioning temperature on the reaction mechanisms. These outcomes were in
335 line with the isothermal calorimetry, strength, porosity and XRD results, as well as earlier
336 observations (Dung and Unluer 2017a), highlighting the influence of moderately elevated
337 temperatures in enhancing the hydration reaction within RMC-based mixes. Out of all the
338 conditions used in this study, pre-conditioning at 50-60°C was identified as the most effective
339 environment to enhance the hydration of MgO, thereby increasing the potential for subsequent
340 carbonation and associated strength gain.

341

342

343 **3.6. Microstructure**

344

345 Fig. 7 shows the SEM images of selected RMC samples after 56 days of curing. The wide-spread
346 formation of rosette-like hydromagnesite was observed in all samples, which aligned with the
347 XRD results. Strength development of RMC samples is related to microstructural evolution as the

348 morphology and the binding strength of the carbonate crystals contribute to the network
349 structure (Liska and Al-Tabbaa 2009, Mo et al. 2017, Ruan and Unluer 2017a, Dung and Unluer
350 2017b, Dung and Unluer 2017a, Dung and Unluer 2018). Accordingly, higher temperatures used
351 in pre-conditioning led to an increase in the crystal size of hydromagnesite in comparison to the
352 control sample (30°C). This increase in the size of these HMC crystals led to denser
353 microstructures, which could contribute to the lower porosity and the improved performance of
354 samples subjected to pre-conditioning under 50-60°C. A similar outcome was also reported in
355 previous studies (Dung and Unluer 2018, Dung and Unluer 2017c), where the effect of carbonate
356 crystal size on the strength of RMC samples was discussed. These results revealed the role of pre-
357 conditioning in not only increasing the reaction rate and degree, but also changing the
358 morphology of the reaction products, which could contribute to the overall performance of RMC
359 samples. Nevertheless, it must be noted that a few SEM images may not provide a sufficient proof
360 of the general distribution of crystal sizes, thereby necessitating further investigation to confirm
361 the changes in crystal size under different temperatures.

362

363

364 **4. Conclusions**

365

366 This study investigated the influence of pre-conditioning on the strength and microstructural
367 development of RMC-based concrete mixes. The obtained results demonstrated the positive
368 influence of subjecting RMC samples to moderately elevated temperatures (50-60°C) for 1-2 days
369 before curing in improving the dissolution of MgO and the precipitation of hydration products.
370 Carbonation was enhanced as a result of an increase in the rate and degree of the hydration
371 reaction. Microstructural analysis revealed that hydromagnesite was the main source of strength
372 in all samples. The amount of hydromagnesite increased in samples subjected to elevated
373 temperatures during pre-conditioning. In addition to the content, the increase in the size of
374 hydromagnesite crystals under higher temperatures could have contributed to improved
375 performance.

376

377 Overall, the findings of this study highlighted the correlation between strength and porosity and
378 reaction mechanisms observed within carbonated RMC-based concrete samples. Accordingly, a
379 reduction in the porosity of RMC samples indicated the densification of the sample
380 microstructure along with the increased formation of hydration and carbonation products, which
381 led to a dense binding network of HMC phases. In this respect, the extent of the carbonation
382 reaction as well as the morphology of the carbonate phases observed in each sample controlled
383 the strength development at different durations. This improved formation of carbonate phases
384 resulted in denser microstructures, thereby facilitating higher strengths in the long-term.

385

386 Within this context, the role of pre-conditioning in enhancing the reaction mechanisms and the
387 associated strength development of carbonated RMC samples was demonstrated. The
388 application of this practical approach to the preparation of RMC-based samples can not only
389 present a more efficient use of the binder component, but also enable the increased
390 sequestration of CO₂ within these samples, thereby lowering the environmental impacts of RMC
391 mixes.

392

393

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395

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398

399

400 **Data Availability Statement**

401

402 e. All data, models, and code generated or used during the study appear in the submitted article.

403

404

405

406 **References**

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