

# 1 Carbonation effect on the properties of lime and cement treated silty and sandy soils

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## 14 Abstract

15 Laboratory investigations on the carbonation of lime and/or cement-treated soils were mostly conducted by  
16 using a carbon-dioxide content up to about 10%, which is much higher than atmospheric carbon-dioxide  
17 ( $\approx 0.03\%$ ). Therefore, the present study examines the physicochemical, pore structure, and compressive  
18 strength evolution of lime and/or cement-treated specimens by exposing the top surface of the specimens  
19 to 0.3% carbon-dioxide content. The study examines carbonation's impact on silty and sandy soils.  
20 Specimens treated with 2.5% quicklime, 5% cement, and a mixture of 1% quicklime and 5% cement  
21 undergo 28 days of carbonation. The strength obtained was maximum for carbon-dioxide exposed cement-  
22 treated soils, followed by combined lime and cement-treated soils, and then lime-treated soils. All carbon-  
23 dioxide exposed specimens showed comparatively greater strength than the corresponding non-exposed  
24 specimens. In addition to calcite deposition, soil suction and cementitious compounds also contributed to  
25 this high strength evolution in the carbon-dioxide exposed specimens. Based on physicochemical and pore  
26 structure analysis, the combined contribution made by soil suction and calcite precipitation towards strength  
27 evolution can be considered almost the same. The significant differences in strength evolution between the  
28 carbonated specimens are attributed to the differences in the development of cementitious compounds. This  
29 development of cementitious compounds is governed by the soil's mineralogy and the binders added. The

30 exposure condition of soil specimens during the carbonation reaction defines the soil suction evolution.  
31 Therefore, the study highlights the importance of reproducing carbon-dioxide content and curing conditions  
32 close to the in-situ situations to accurately assess the strength evolution of carbonated specimens.

33 Keywords: Carbon-dioxide content; unconfined compressive strength; soil suction; pore structure; lime  
34 and/or cement-treated soils.

## 35 **Introduction**

36 Soil management is a critical challenge in any land development project of geotechnical engineering. Thus,  
37 the process of soil improvement is a practice that is widely implemented to make use of the soil located  
38 directly on the worksite. Soil improvement involves improving the engineering properties of the soil, both  
39 chemically and physically. Physical improvement consists of the implementation process, such as mixing  
40 soil and water with binders, compaction, etc. (Cuisinier et al. 2011; Das et al. 2021; Makki-Szymkiewicz  
41 et al. 2015; Runigo et al. 2009; 2011), while chemical improvement includes the use of organic or inorganic  
42 binders (Baghdadi et al. 1995; Chethan and Ravi Shankar 2021; Koliasset al. 2005; Miller and Azad 2000;  
43 Show et al. 2003; Wang et al. 2022).

44 Of the several conventional chemical binders used, lime and cement are often used to enhance the  
45 long-term performance of soils (Ho et al. 2017; 2018; Mahedi et al. 2020; Nakarai and Yoshida 2015). The  
46 basic chemical reactions remain the same when lime and cement come into contact with a soil-water mixture  
47 (Bhattacharja et al. 2003; Diamond and Kinter 1965; Ho et al. 2017). However, the extent of these reactions  
48 varies with whether the binder added is lime or cement (Bhattacharja et al. 2003).

49 Given that lime and cement treatment improve the performance of soils, several factors must be  
50 considered while concluding the overall evolution of these treated soils. Such a factor involves the  
51 compaction conditions implemented during construction (Das et al. 2020; 2021); the content of binder  
52 added (Bergado et al. 1996; Lemaire et al. 2013); the soil type (Azadegan et al. 2012); the curing conditions  
53 (Lemaire et al. 2013) etc. At the laboratory scale, the curing condition often involves the process of  
54 wrapping a soil sample and then curing it at a controlled ambient temperature and relative humidity. Such

55 a process prevents the specimen from being exposed to an environment that soil is usually known to  
56 encounter in the field. In the field, along with the stress effect generated due to changes in temperature and  
57 relative humidity on exposed treated soil, the occurrence of carbonation was reported in several studies  
58 (Das et al. 2022c; Deneele et al. 2021; Kleib et al. 2024; Padmaraj and Arnepalli 2021).

59         The impact of the carbonation reaction on the chemically modified lime and cement-treated soils  
60 in terms of physical, chemical, and microstructural properties is investigated to some extent (Das et al.  
61 2022c; Deneele et al. 2021; Ho et al. 2017; 2018; Nakarai and Yoshida 2015; Padmaraj and Arnepalli 2021).  
62 For instance, Nakarai and Yoshida (2015) compared the effect of natural and 5% accelerated CO<sub>2</sub> content  
63 on the strength evolution of 8% cement-treated silica sand. They concluded that due to the carbonation of  
64 the cement-treated sand, the formation of CaCO<sub>3</sub> occurred, which is the main reason for the long-term  
65 strength evolution of the sand. The porous structure of cement-treated sand enhanced the deeper penetration  
66 of CO<sub>2</sub> gas. Ho et al. (2017) investigated the carbonation effect on the strength evolution of cement-treated  
67 sand mixtures and sand-loam mixtures. Based on the investigation, the change in water content during  
68 carbonation is demonstrated to play a significant role in the strength increase of cement-treated sand.  
69 Another study by Ho et al. (2018) showed that an increase in the internal water content of cement-treated  
70 soil limits carbonation. Deneele et al. (2021) used 10% CO<sub>2</sub> content to evaluate the effect of carbonation  
71 on the Unconfined Compressive Strength (UCS) and microstructure of lime-treated silty soil. The study  
72 demonstrated the carbonation of C-(A)-S-H gel to be detrimental to strength evolution by evidencing the  
73 change in soil fabric and pore-structure evolution during carbonation. Another study by Padmaraj and  
74 Arnepalli (2021) provided extensive insight into the carbonation effect on the fabric and pore structure of  
75 4% and 8% lime-treated silty clay soil. This study demonstrated the negative effect of short-term and long-  
76 term carbonation on Unconfined Compressive Strength evolution by eliminating suction and water content  
77 evolution during carbonation. Das et al. (2022c) demonstrated a field investigation highlighting the  
78 occurrence of carbonation depth based on microstructural studies in specimens sampled from a 2.5% lime-  
79 treated silty soil experimental embankment after one year of atmospheric exposures. Another field  
80 investigation was made by Haas and Ritter (2019), who studied a 2.5% quicklime-treated embankment after

81 11 years and 34 years from treatment. They reported a rise in UCS between 2.8 and 3.4 MPa after 11 years,  
82 and the UCS further increased up to 6 MPa after 34 years from treatment. Of the total lime added during  
83 construction, 37% is used in carbonation, 47% in pozzolanic reactions, and 16% free CaO is still available.  
84 From their study, they proposed that a further increase in UCS can be predicted. While the above studies  
85 mainly focussed on the carbonation mechanism and its impact on strength evolution, a recent study was  
86 made on the optimization of carbonation reactions of lime-treated soils by Kleib et al. 2024. Kleib et al.  
87 (2024) optimized the occurrence in carbonation rate of 1, 2, and 4% quicklime treated silty soil. They stated  
88 that even specimens submitted to 0% CO<sub>2</sub> showed a higher carbonation rate. The phenomenon was  
89 attributed to the early occurrence of carbonation, particularly during the mellowing period of the specimens  
90 before compaction.

91 Thus, some of the existing laboratory studies using a carbonation chamber demonstrated the  
92 occurrence of carbonation as the sole responsible for the rise of strength in cement-treated sandy soil, while  
93 other parts described carbonation as a negative effect on the pozzolanic compounds and the strength  
94 evolution of lime-treated soil. Again, field investigation shows a rise in UCS in the lime-treated soil even  
95 after carbonation. Moreover, the mellowing duration of soil before subjecting them to anticipated CO<sub>2</sub>  
96 content was also shown to have a significant impact on the ultimate outcome of carbonation impact  
97 observed in terms of mechanical performance and carbonation rate. In conclusion, the existing literature  
98 provides a varied perspective on the effects of carbonation on lime and cement-treated soils. Thus,  
99 additional studies on the carbonation impact of lime and/or cement-treated soils are essential. The intention  
100 of the use of a carbonation chamber at the laboratory scale is to reproduce the situation a treated soil is  
101 likely to experience when exposed to atmospheric CO<sub>2</sub> at the existing field temperature. Generally, a CO<sub>2</sub>  
102 content of 0.03% is reported as atmospheric CO<sub>2</sub> (Xu et al. 2022). However, most of the studies injected a  
103 CO<sub>2</sub> content of up to about 10% to study accelerated results, which is much higher than the actual content  
104 treated soil encounters in the field. Also, little is reported regarding the evolution of soil suction and water  
105 content during such studies.

106 Thus, in the wake of the above context, the existing study presents an evaluation of the carbonation  
107 effect on silty and sandy soil treated with lime, cement, and a mixture of lime and cement. The study aims  
108 to expose the top region of the treated specimens to 0.3% CO<sub>2</sub> content, a content about 10% higher than the  
109 atmospheric level, in order to maintain a slightly accelerated reaction. The evolution of carbonation depth  
110 and the UCS, physicochemical, and microstructural properties of the carbonated soil are evaluated. Thus,  
111 this study details the observations made on lime and/or cement-treated soil properties by exposing the top  
112 region of the soil to a CO<sub>2</sub> content slightly higher than the one available in in-situ situations but much lower  
113 than the one often practiced at the laboratory. It is worth noting that the impact of the possible occurrence  
114 of carbonation in the soil during mixing is ignored herein. To gain a wider insight, the study was conducted  
115 using two different soil types. The first part of the study explains the carbonation effect on soil pH and how  
116 the pH alteration caused pore modifications. Then, the contribution of pore modifications and soil suction  
117 toward UCS evolution is discussed.

## 118 **Materials**

119 Two different soils supplied by different company's contractors were used in this study. One soil is  
120 categorized as A2, *i.e.*, silty soil, and the other as B4/B5, *i.e.*, sandy soil, as per the European norm NF P11-  
121 300 (1992). As per the USCS standard A2 soil stands for inorganic clays, and B4/B5 soil stands for silty  
122 sands. A2 soil was imported from Obourg, Belgium, and B4/B5 was imported from Versailles, France.

123 Both quicklime (CaO) and cement, supplied by different commercial suppliers, were used as  
124 binders to evaluate the difference in carbonation effect in different soils treated with different binders. The  
125 quicklime used has been previously reported in studies by Das et al. (2020; 2021; 2022a; 2022b) and  
126 Ranaivomanana et al. (2022). Lime consists of 90.9% of available CaO and a reactivity ( $t_{60}$ ) of 3.3 min.  
127 The cement is composed of 98% clinker and has a mechanical strength of 2 MPa at 2 days and 65 MPa at  
128 28 days. It is an accelerated reactivity cement.

129 The Lime Modification Optimum (LMO) of the soil was determined as per ASTM D 6276-99a  
130 (2006). The LMO obtained was 1% for Obourg's soil and 0.5% for Versailles's soil.

131

## 132 **Sample preparation and Laboratory tests**

133 The compaction characteristics of the untreated Obourg (OB) and Versailles (VR) soils were determined  
134 by the standard Proctor compaction test as per ASTM D698-12e2 (2012).

135 The two soils were air-dried and then sieved through a 5 mm sieve. They were then mixed at the  
136 targeted water content. The soil paste was then left in sealed bags for about 24 hours to attain uniformity in  
137 moisture distribution. Then, the soil paste was mixed with quicklime and cement in different proportions  
138 for compaction. They are as follows: Specimens were prepared with (i) 2.5% quicklime (L), *i.e.*, at a lime  
139 content higher than LMO, to ensure the availability of sufficient lime for the long-term pozzolanic  
140 reactions; (ii) with the combination of 1% quicklime (L) and 5% cement (C). The use of 5% cement is  
141 prescribed as the economic cement quantity to be added to a given soil in France; (iii) with 5% cement (C).  
142 The types of soil paste-binder mixtures prepared are listed in Table 1.

143 Soil mixtures were then rested for 1 hour before kneading compaction. The process of kneading  
144 compaction involves the application of a dynamic load successively using a 3-feet kneading tool at an angle  
145 of 45° between two successive loadings. Additional details regarding kneading compaction can be obtained  
146 in the study reported by Das et al. (2021).

147 A total of 48 cylindrical specimens of dimensions having a length of 10cm and a diameter of 5cm  
148 were prepared by kneading compaction. Each compacted specimen was then subjected to three different  
149 curing conditions, which are presented in Table 2, along with the total number of specimens and specimen  
150 ID. Specimens were cured in the climatic chamber and carbonation chamber. A temperature of 20°C  
151 (ambient laboratory temperature) and a relative humidity (RH) of 65% are maintained in both chambers.  
152 This selected RH at 20°C was shown to induce maximum carbonation in concrete (Verbeck 1958). Table 2  
153 shows that additional specimens were prepared for each soil configuration. For specimens subjected to 0-  
154 day curing, 2 duplicate specimens were prepared, each subjected to UCS test. For specimens subjected to

155 28-days curing, 3 duplicate specimens were prepared, two of which were subjected to UCS test and the  
156 remaining for physicochemical and microstructure evaluation.

157 For curing in the climatic chamber, specimens were wrapped on all sides, while specimens cured  
158 in the carbonation chamber were left open at the top to allow access to CO<sub>2</sub> in the soils. Fig. 1a shows a  
159 wrapped specimen prepared for curing in the carbonation chamber. The specimens were preferred to be  
160 exposed to CO<sub>2</sub> only on the surface to represent the surface exposure of in-situ treated soil infrastructures.  
161 It is worth noting that this study focuses on the carbonation development in samples cured within the  
162 carbonation chamber, compared to control samples cured in a climatic chamber. Thus, specimens cured in  
163 the climatic chamber were the reference specimens. Consequently, any potential carbonation that might  
164 have taken place during sample preparation in an open laboratory environment is disregarded.

165 No curing specimens and specimens subjected to 28 days of curing were then subjected to the UCS  
166 test, which involves the application of a load sensor at a constant displacement rate of 1mm/min.

167 The remaining duplicate specimens at the end of curing time were subjected to water content, soil  
168 suction, pH, X-ray diffraction (XRD), and pore structure analysis. Measurement of water content involves  
169 the use of the oven drying method as per ASTM D2216-10 (2010), and soil suction was determined using  
170 a WP4C Dewpoint Potentiometer. For soil pH analysis, specimens were crushed and mixed with water in  
171 a liquid soil ratio of 5:1 for 1 hour, and then the pH of the soil was recorded as per ASTM, D4972-19  
172 (2019). XRD and pore structure analysis were performed on freeze-dried specimens. XRD was analyzed  
173 using a PAalytical X'Pert pro MPD diffractometer (Panalytical, Malvern, UK) equipped with a PIXcel 1D  
174 detector (active length of 3.347°2θ). Additional details regarding the test can be obtained from the study  
175 reported by Poussardin et al. (2023). Both Mercury Intrusion Porosimetry (MIP) and Brunauer–Emmett–  
176 Teller (BET) tests were used to observe a complete range of pore modifications. The processes of MIP and  
177 BET tests can be referred to in Romero and Simms (2008) and Westermarck (2000), respectively.

178 The analysis of the above physicochemical properties and pore structure modifications was made  
179 in specimens sampled carefully from the core for the specimens cured in the climatic chamber. However,  
180 for specimens cured in a carbonation chamber, these analyses varied based on the requirements of the study.

181 Fig. 1b presents different sections of the specimens sliced after curing in a carbonation chamber and then  
182 selected for pH, water content, soil suction, and pore structure analysis. The reason for such selection is  
183 highlighted in the results and discussion section.

184

## 185 **Results and discussions**

### 186 *Evolution of soil pH*

187 The pH of soil upon treatment with lime and cement increases to a value greater than 12, owing to the  
188 release of OH<sup>-</sup> ions in the pore water (Bell 1996; Diamond and Kinter 1965; Xu et al. 2020). This evolution  
189 was confirmed in the present treated specimens, as seen in Fig. 2. The pH of the untreated OB and VR  
190 soils was measured to be about 8.20. This pH increased to a value somewhere between 12 to 12.5 for both  
191 OB and VR treated and 28-day climatic chamber cured specimens.

192 To observe the impact of carbonation on soil pH with respect to the subjected specimen's depth,  
193 i.e., carbonation depth, specimens were sampled from the top, middle, and bottom as specified in Fig. 1b.  
194 Fig. 2a and b present the difference in pH measured from specimens sampled at different depths for OB  
195 and VR, respectively.

196 Compared to the specimens cured in the climatic chamber, the pH level is relatively lower in all  
197 the top-layered specimens sampled from specimens cured in the carbonation chamber. However, the pH  
198 value recorded in the middle and bottom layers of the sampled specimens was similar to the one obtained  
199 from the climatic chamber-cured specimens. This difference in pH level measured between the bottom and  
200 middle sampled specimens and the top specimen probably indicates the occurrence of carbonation depth  
201 within 2cm from the surface during the 28 days of curing at the carbonation chamber. This is confirmed in  
202 the following studies.

203

204



205 ***XRD analysis***

206 Using XRD analysis, differences in the presence of minerals between specimens treated with different  
207 binders and cured under different conditions are presented along with the respective untreated specimens  
208 in Fig. 3. The preceding study confirms the alteration of soil pH in the top sampled specimens of the  
209 specimens cured at the carbonation chamber; hence, only these specimens were subjected to XRD analysis.

210 Fig. 3a shows the presence of kaolinite, illite, chlorite, and muscovite as clay minerals, orthoclase  
211 and albite as feldspar minerals, and quartz in the untreated OB specimen. A quite similar intensity and  
212 position of these minerals were also identified in all the treated and 28-day climatic chamber cured OB  
213 specimens. For all treated specimens cured in the carbonation chamber, a new peak at 2theta equal to about  
214 29.5° is observed. This observation suggests the presence of calcite. Thus, the presence of calcite is  
215 confirmed in the top-sampled specimens cured in the carbonation chamber.

216 For raw VR specimens, kaolinite and illite as clay minerals, orthoclase as a feldspar mineral, and  
217 quartz are observed to be present (Fig. 3b). In all the treated VR specimens, in addition to the preceding  
218 mineral's presence, of Portlandite is observed at 2theta equal to about 18°. Similar to OB, VR carbonation  
219 chamber-cured specimens also showed the presence of calcite. Thus, carbonation of the top-sampled  
220 specimens is confirmed for both OB and VR treated, and carbonation cured specimens through XRD  
221 analysis.

222 For the convenience of understanding, top-sampled specimens for specimens cured in the  
223 carbonation chamber are referred to as carbonated specimens, while the corresponding specimens cured in  
224 the climatic chamber are referred to as non-carbonated specimens in the following study. However, the  
225 representation in the figures is kept as per the specimen ID presented in Table 2 to maintain uniformity in  
226 the legends of all figures.

227 ***Evolution of pore-structures in treated soils***

228 Previous studies showed that lime and/or cement-treated soils show a decrease in the available larger pores  
229 and lead to the generation of smaller pores, mostly of a diameter smaller than 3000 Å (Das et al. 2021;

230 Horpibulsuk et al. 2010; Khalid et al. 2018). A complete modification of these pore networks can be  
231 assessed elaboratively using both the MIP and BET tests (Das et al. 2020). MIP was reported to be  
232 preferable to observing the larger pores, while BET was for smaller pores (Das et al. 2020). Thus, in this  
233 study, using MIP, pores greater than 3000 Å are analyzed, while pores lower than 3000 Å are analyzed by  
234 the BET test.

### 235 ***Pore-structure observation by MIP***

236 Fig. 4 presents the pore structure of the untreated and treated non-carbonated and carbonated OB and VR  
237 specimens. Since the XRD analysis confirmed the occurrence of carbonation in the top-sampled specimens  
238 by evidencing the calcite presence, only these specimens were subjected to the MIP test and the remaining  
239 tests, which were discussed in the following sections.

240 Untreated OB soil showed the availability of highly intense, large pores of diameter  $1.5 \times 10^4$  Å.  
241 Regardless of the type of binders added, the large pores observed in untreated specimens were no longer  
242 available in the non-carbonated specimens (Fig. 4a-c). Instead, the development of pores smaller than 3000  
243 Å was observed. Carbonated OB specimens exhibited a low intense pore peak at diameter  $1 \times 10^4$  Å and a  
244 small number of pores smaller than 3000 Å.

245 Untreated VR soils showed a pore peak at  $2 \times 10^5$  Å, which is larger as well as of much higher  
246 intensity compared to the one observed in the corresponding untreated OB soil (Fig. 4d-f). Unlike the OB  
247 soil, the non-carbonated VR soil showed a difference in pore modifications based on the binders added. L-  
248 treated VR soil exhibits 3 different pore peaks. A narrow peak at  $2 \times 10^5$  Å, which is similar to the one  
249 available in untreated VR, but of lower intensity, another small peak at  $4 \times 10^4$  Å, and a small intensity  
250 broad peak over pores smaller than 3000 Å (Fig. 4d). For L + C and C-treated VR specimens, the preceding  
251 narrow peak at  $2 \times 10^5$  Å was missing; however, pores at  $4 \times 10^4$  Å, and pores smaller than 3000 Å were  
252 observed (Fig. 4e & f).

253 Similar to the L-treated VR non-carbonated specimen, the corresponding carbonated soil showed  
254 a narrow pore peak at  $2 \times 10^5$  Å (Fig. 4d). Additionally, a distinct pore peak at  $7 \times 10^4$  Å was observed, and

255 a negligible number of pores smaller than 3000 Å. Pore structures of L + C and C-treated VR carbonated  
256 soils exhibited a distinct peak at  $7 \times 10^4$  Å and some number of pores smaller than 3000 Å (Fig. 4e & f).

257

### 258 ***Pore-structure observations by BET***

259 Fig. 5 presents the presence of pores smaller than 3000 Å analyzed by the BJH method from the results  
260 obtained by the BET test. Both untreated OB and VR soils showed a pore peak at 40 Å, which is attributed  
261 to the presence of clay porosity (Das et al. 2020).

262 Compared to OB untreated specimens, OB non-carbonated specimens showed a significant  
263 evolution of smaller pores over the pore range of 50 to 2000 Å (Fig. 5a-c). Such an evolution was significant  
264 in the C-treated soil (Fig. 5c), followed by L + C-treated (Fig. 5b) and L-treated (Fig. 5a) soils. However,  
265 OB L+C and C-treated carbonated specimens also showed the development of pores over the pore range  
266 50 to 2000 Å, but at comparatively lower intensities than the non-carbonated specimens. This evolution  
267 was almost negligible in the OB L-treated carbonated specimen (Fig. 5a).

268 The overall trend observed in the generation of pores smaller than 2000 Å remained the same in  
269 the corresponding VR specimens (Fig. 4d-f). The discussions related to the observations made in MIP and  
270 BET tests are presented separately in the next section to provide an integrated understanding of the pore  
271 modifications.

272

### 273 ***Summary of the observed pore structure evolutions***

274 Given that OB is silty soil and VR is sandy soil, an order of magnitude larger pore size and greater intensities  
275 are expected in the VR than in the OB untreated soil (Fig. 4).

276 Due to treatment and 28 days of curing, precipitation of cementitious compounds has occurred,  
277 which resulted in the development of pores smaller than 3000 Å (Das et al. 2021; 2022a). This development  
278 of smaller pores was observed to be dependent on the soil type, and the binder added.

279 Irrespective of the type of treatment, OB non-carbonated soil, at the end of curing, showed a total  
280 decrease of the larger pores observed in the corresponding untreated specimen (Fig. 4a-c). This observation  
281 remained the same in the corresponding VR specimens (Fig. 4e & f), except for the VR L-treated and non-  
282 carbonated soil (Fig. 4d). Such a difference indicates that 2.5% lime treated VR soil after 28 days of curing  
283 time showed the generation of smaller pores (Fig. 5d); however, the relative transformation of the larger  
284 pores to smaller pores owing to the precipitation of cementitious compounds is lower than the specimens  
285 subjected to L+C and C treatment (Fig. 5e & f). Again, all types of non-carbonated VR specimens showed  
286 a peak at around  $4 \times 10^4 \text{ \AA}$ , along with the generation of pores smaller than  $3000 \text{ \AA}$ . On the other hand, all  
287 corresponding OB soil shows the only generation of pores smaller than  $3000 \text{ \AA}$  and of relatively greater  
288 intensities. These differences in the kinetics of the development of smaller pores in OB and VR soils may  
289 be attributed to the presence of greater intensities of clay fraction in OB than in VR soil, as evident from  
290 Fig. 3. The presence of relatively greater intensities of clay fraction probably has caused greater availability  
291 of pozzolanas in the OB soil, which might have led to the higher development of cementitious compounds.  
292 Again, the availability of Portlandite in the VR non-carbonated soil during XRD analysis (Fig. 3b) indicates  
293 a limited occurrence of pozzolanic reactions, which again supports the relatively low development of  
294 pozzolanic compounds in the VR non-carbonated soil.

295 However, for all carbonated OB-treated specimens, the narrow pore peak at  $1.5 \times 10^4 \text{ \AA}$  was  
296 lowered, giving rise to new low-intensity pores at  $1 \times 10^4 \text{ \AA}$  (Fig. 4a-c) and a small number of pores smaller  
297 than  $3000 \text{ \AA}$  (Fig. 5a-c). This indicates that a part of the large pores at  $1.5 \times 10^4 \text{ \AA}$  gave rise to pores smaller  
298 than  $3000 \text{ \AA}$  due to the development of cementitious compounds, while the new pore peak at  $1 \times 10^4 \text{ \AA}$  can  
299 be attributed to the deposition of calcite. The deposition of calcites was reported to show greater pores than  
300 those observed from the deposition of pozzolanic compounds (Deneele et al. 2021). Deneele et al. (2021)  
301 reported that, owing to the transformation of pozzolanic compounds into  $\text{CaCO}_3$  rhombohedra (probably  
302 calcite), an increase in the mean size of the existing modified pores occurred. In VR-L+C and C-treated  
303 carbonated specimens, the large pore peak at  $2 \times 10^5 \text{ \AA}$  observed in the untreated soil disappeared and gave  
304 rise to a distinct pore peak at  $7 \times 10^4 \text{ \AA}$  (Fig. 4e & f), and a small number of pores smaller than  $3000 \text{ \AA}$

305 (Fig. 5e &f). While the latter was due to cementitious compound deposition, the former can be attributed  
306 to calcite precipitation. This behavior was slightly different for the VR-L treated carbonated soil, where,  
307 along with pores at  $7 \times 10^4 \text{ \AA}$ , a pore peak at  $2 \times 10^5 \text{ \AA}$  was also visible, but of lower intensities than the  
308 untreated soil. Thus, VR-L treated carbonated specimens showed calcite deposition pores; however, pores  
309 smaller than  $3000 \text{ \AA}$ , which were visible for the other VR-carbonated specimens, remain negligible  
310 compared to the ones observed for the corresponding non-carbonated specimens. The difference in the pore  
311 peak indicating the precipitation of calcite was  $7 \times 10^4 \text{ \AA}$  for VR, while it was  $1 \times 10^4 \text{ \AA}$  for OB. With the  
312 availability of greater intensities of large pores in VR untreated soil (Fig. 4), precipitation of calcites has  
313 resulted in pores greater than those observed for OB carbonated soil.

314         The above evolution of pores smaller than  $3000 \text{ \AA}$  anticipated in all types of treated soils was of  
315 relatively greater intensities in C-treated OB and VR soils, followed by L + C and then L-treated soils (Fig.  
316 5). Such an occurrence is expected owing to the immediate formation of C-S-H, a cementitious compound,  
317 upon hydration of Portland cement (Bhattacharja et al. 2033; Consoli et al. 2009; 2011). Meanwhile, in  
318 soil-lime reactions, C-S-H is formed in the long-term reactions upon the availability of calcium from lime,  
319 pozzolans, and water in the soil. However, the L+C-treated specimen showed comparatively lower  
320 intensities of these smaller pores than the C-treated specimen, particularly in OB soil (Fig. 5b & c). Such a  
321 phenomenon is probably due to the lower availability of water during cement addition. During the  
322 preparation of L+C-treated soil, after adding 2.5% lime to the soil paste, 5% cement was added. Since the  
323 addition of lime has used up some of the available water due to the exothermic reactions, a low amount of  
324 water can be said to have been available for the hydration reaction upon cement addition, thus leading to  
325 the availability of more hydrates in C-treated soils than in L+C-treated soils.

326

### 327 ***Evolution of hydric parameter of the soil: soil suction and water content***

328 Since specimens exposed to  $\text{CO}_2$  were unwrapped on top (Fig. 1a), while the corresponding climatic  
329 chamber-cured specimens were fully wrapped, the suction generation during curing will differ based on

330 these curing conditions. The difference in the evolution of average soil suctions of the carbonated and non-  
331 carbonated specimens and the corresponding water content loss with respect to initial water content is  
332 presented in Fig. 6.

333 As expected, owing to the exposure of the top of the specimens in the carbonated chamber, during  
334 curing, a significant water content of up to about 85% was lost with respect to the initial water content (Fig.  
335 6b). This obviously led to the notifiable suction evolution of about 16 MPa in the carbonated specimens,  
336 which remains negligible in the corresponding non-carbonated specimens (Fig. 6a). Some of the water  
337 content that was observed to be lost with respect to the initial specimens in the non-carbonated is attributed  
338 to the consumption of water during pozzolanic reactions (Fig. 6b). However, the trend in this evolution of  
339 suction and water content loss varied with the soil type and the binder added to the carbonated specimens.  
340 This is probably due to certain chemical modifications, which will be evaluated in further studies, and are  
341 not discussed herein since the scope of the suction evaluation is to observe its contribution towards strength  
342 evolution, as discussed in the next section.

343

#### 344 ***Evolution of UCS in treated soils***

345 The evolution of UCS in terms of stress-strain curves for the non-carbonated and carbonated  
346 specimens is compared with the 0-day cured treated OB and VR specimens in Fig. 7. The presentation was  
347 made for all duplicate specimens to ensure the accuracy of the evolution. The average UCS values of the  
348 presented specimens are provided in Table 3.

349 OB soil immediately after treatment, i.e., under no curing conditions, showed a UCS level in the  
350 range of 0.17-0.22 MPa, which is almost similar regardless of the type of binder added to the soil. This  
351 behavior was also observed with the VR-treated specimens, where the UCS level remained at 0.05 MPa  
352 (Table 3). However, the UCS level obtained with the OB soils was relatively higher than the corresponding  
353 VR soils. Owing to a greater clay fraction in OB soil (Fig. 3), a greater soil agglomeration can be expected,  
354 which might have resulted in this relatively greater UCS evolution.

355           After 28 days of curing in a climatic chamber, the UCS increased in both OB and VR non-  
356 carbonated specimens. This is due to the development of smaller pores (Fig. 4 & 5) owing to the  
357 precipitation of cementitious compounds, which agrees with previous findings (Consoli et al. 2009; Das et  
358 al. 2020). However, this increase in UCS was relatively lower in all corresponding VR-non-carbonated  
359 specimens. Such an observation is attributed to the greater intensities of smaller pores development in the  
360 OB specimens compared to the one observed in the corresponding VR specimens (Fig. 5). The trend of this  
361 UCS was maximum for C treated, intermediate for L+C treated, and minimum for L treated OB and VR  
362 non-carbonated specimens. Such an occurrence is due to the greater availability of pores smaller than 3000  
363 Å in C-treated OB and VR soils than in L + C and L-treated soils (Fig. 5). A greater generation of smaller  
364 pores results in the observed greater strength (Das et al. 2021).

365           However, all carbonated specimens showed a level of UCS higher than the non-carbonated  
366 specimens. OB carbonated specimens showed 0.30, 0.70, and 1.38 MPa higher average UCS in L, L+C,  
367 and C treated than the UCS obtained with the corresponding non-carbonated soils (Fig. 7a-c). This behavior  
368 was also observed in VR specimens. However, it is insignificant compared to the one observed in OB  
369 carbonated specimens. The increased UCS level in the carbonated specimens is attributed to the combined  
370 contribution made by the high average suction evolution (Fig. 6a), calcite precipitation (Fig. 3), as well as  
371 smaller pores development (Fig. 4&5). Das et al. (2023) have stated how increased suction due to  
372 considerable water content loss can lead to increased UCS in lime-treated soil. Owing to the quick  
373 precipitation of calcites in soil pores, which enhances soil resistance, calcite development was shown to  
374 increase the strength of treated soil at a greater rate during short-term curing (Dimitri et al. 2021).  
375 Considering that the combined contribution made by soil suction and calcite precipitation towards strength  
376 evolution in OB and VR carbonated specimens is almost the same, the significant difference in UCS  
377 evolution in these carbonated specimens can be attributed to the difference in the development of  
378 cementitious compounds between these two soil types. This development of cementitious compounds is  
379 governed by the soil types and the binders added, as explained before.

380

## 381 **Conclusions**

382 This study is focused on observing the carbonation evolution under 0.3% of CO<sub>2</sub> content and its effect on  
383 the UCS evolution of 28 days cured lime, cement, and combined lime and cement-treated silty and sandy  
384 soils. The observation is discussed based on the evaluation of soil's pH, suction, and pore structure  
385 modifications. The following are the conclusions derived from the study:

- 386 1) After 28 days of carbonation, the maximum pH decrease was only up to 10.8 from 12.5 in the lime-  
387 treated and carbonated OB soil. The decrease in pH level observed in the cement and combined lime  
388 and cement-treated carbonated OB and VR soils remained in between 11 to 12.3. Owing to the  
389 exposure of specimens to a CO<sub>2</sub> content of only 0.3%, slow calcite formation occurred, thus leaving  
390 enough OH<sup>-</sup> ions, which resulted in the formation of cementitious compounds along with calcites in  
391 the carbonated samples.
- 392 2) Alongside the development of calcite in the carbonated samples, the development of cementitious  
393 compounds also occurred, as reflected by the presence of pores smaller than 3000 Å. This significantly  
394 contributed to the higher UCS level observed in the carbonated soils. Based on the soil types and  
395 binders added, the development of cementitious compounds varied. OB soil having higher pozzolanas  
396 due to a greater presence of clay fraction showed higher, smaller pores evolution. Cement-treated soil  
397 shows the highest UCS values compared to lime-treated and combined lime and cement-treated soils,  
398 owing to the immediate formation of C-S-H due to hydration.
- 399 3) Suction evolution, which occurred during carbonation, owing to the exposure of the top region of the  
400 samples during curing, significantly contributes to the high UCS level observed in all the treated  
401 specimens after 28 days of carbonation. This condition of soil curing represents the surface exposure  
402 of in-situ treated soil infrastructures, where surface carbonation is highly possible. Thus, emphasis  
403 should be laid on the curing conditions reproduced at the laboratory scale while predicting the UCS  
404 evolution in in-situ carbonated samples.



405 4) During carbonation, the availability of pozzolanas in the subjected soil types and the types of binder  
406 added define the intensities of the deposition of both calcites and cementitious compounds, which  
407 contribute towards the UCS evolution. The UCS of OB-treated and carbonated soil, particularly for  
408 the one subjected to cement treatment, is almost three times higher than the corresponding VR soil.

409 Thus, the study highlights that the carbonation of lime-, cement- and combined lime and cement-treated  
410 soils under 0.3% CO<sub>2</sub> content develops not only calcites but also cementitious compounds. The study also  
411 reflects the importance of curing conditions that need to be reproduced during carbonation studies, as this  
412 will affect the evolution of soil suction. In this study, the development of soil suction, calcites, and  
413 cementitious compounds together brings a UCS level that is higher in carbonated soils than in non-  
414 carbonated soils. Hence, importance should be given to the laboratory reproduction of CO<sub>2</sub> content and the  
415 curing conditions close to the actual in-situ situations. The present study reports the carbonation effect for  
416 28 days; hence, future studies should be conducted on the long-term carbonation effect to understand if the  
417 impact of carbonation remains the same or different from the present investigations.

#### 418 **Data Availability Statement**

419 Data generated or analyzed during the study is included in the submitted manuscript.

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425

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566 **Table 1.** Soil paste and binder mixtures prepared for compaction.

Soils	Mixtures
	OB + 2.5% L
Obourg (OB)	OB + 1% L + 5% C
	OB + 5% C
	VR + 2.5% L
Versailles (VR)	VR + 1% L + 5% C
	VR + 5% C

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581 **Table 2.** Type of curing conditions implemented, specimen ID, and number of specimens prepared.

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Curing conditions	Soil mixtures	Specimens ID	Number of specimens
0-day curing/no curing	OB + 2.5% L	OB-L-0d	2
	OB + 1% L + 5% C	OB-L + C-0d	2
	OB + 5% C	OB-C-0d	2
	VR + 2.5% L	VR-L-0d	2
	VR + 1% L + 5% C	VR-L + C-0d	2
	VR + 5% C	VR-C-0d	2
28-days curing at climatic chamber	OB + 2.5% L	OB-L-28d	3
	OB + 1% L + 5% C	OB-L + C-28d	3
	OB + 5% C	OB-C-28d	3
	VR + 2.5% L	VR-L-28d	3
	VR + 1% L + 5% C	VR-L + C-28d	3
	VR + 5% C	VR-C-28d	3
28-days curing at carbonation chamber	OB + 2.5% L	OB-L-28d-CO <sub>2</sub>	3
	OB + 1% L + 5% C	OB-L + C-28d-CO <sub>2</sub>	3
	OB + 5% C	OB-C-28d-CO <sub>2</sub>	3
	VR + 2.5% L	VR-L-28d-CO <sub>2</sub>	3
	VR + 1% L + 5% C	VR-L + C-28d-CO <sub>2</sub>	3
	VR + 5% C	VR-C-28d-CO <sub>2</sub>	3

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590 **Table 3.** Average UCS values of specimens observed in Fig. 7

Situation	OB soils		VR soils	
	Specimens ID	Average UCS (MPa)	Specimens ID	Average UCS (MPa)
No curing	OB-L-0d	0.22	VR-L-0d	0.05
Cured in climatic chamber	OB-L-28d	0.50	VR -L-28d	0.36
Cured in CO <sub>2</sub> injected chamber	OB-L-28d-CO <sub>2</sub>	0.76	VR -L-28d-CO <sub>2</sub>	0.38
No curing	OB-L+C-0d	0.20	VR -L+C-0d	0.05
Cured in climatic chamber	OB-L+C-28d	1.49	VR -L+C-28d	0.79
Cured in CO <sub>2</sub> injected chamber	OB-L+C-28d-CO <sub>2</sub>	2.19	VR -L+C-28d-CO <sub>2</sub>	1.04
No curing	OB-C-0d	0.17	VR -C-0d	0.05
Cured in climatic chamber	OB-C-28d	1.82	VR -C-28d	0.90
Cured in CO <sub>2</sub> injected chamber	OB-C-28d-CO <sub>2</sub>	3.20	VR -C-28d-CO <sub>2</sub>	1.12

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601 **Figure Captions List**

602 **Fig. 1.** Picture showing wrapping of specimens prepared for curing in carbonation chamber (a) and  
603 specimen sliced as different depths for physicochemical and pore structure analysis

604 **Fig. 2.** Comparative evolution of pH measured after 28 days of curing in climatic and carbonation chambers  
605 in OB (a) and VR (b) L, L+C, and C-treated soils

606 **Fig. 3.** Comparative XRD analysis between untreated and L, L+C, C-treated OB (a) and VR (b) soils after  
607 28 days of curing at climatic and carbonation chambers.

608 **Fig. 4.** Comparative pore structure evolution in untreated, (a) L, (b) L+C, (c) C-treated OB and untreated  
609 (d) L, (e) L+C, (f) C-treated VR noncarbonated and carbonated specimens by MIP test.

610 **Fig.5.** Comparative pore structure evolution in untreated, (a) L, (b) L+C, (c) C-treated OB and untreated  
611 (d) L, (e) L+C, (f) C-treated VR noncarbonated and carbonated specimens by BET test.

612 **Fig.6.** Comparative evolution in average soil suction (a) and corresponding loss in water content (b) of non-  
613 carbonated and carbonated OB and VR specimens.

614 **Fig.7.** Stress-strain evolution in OB (a-c) and VR (d-f) treated and non-carbonated, treated, and carbonated  
615 specimens compared to treated and 0 days cured specimens.