

CO2 Foamed Concrete: An Innovative Approach to Combining Mechanical Strength and Carbon Storage

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1                   **CO<sub>2</sub> Foamed Concrete: an innovative approach to combining**  
2                   **mechanical strength and carbon storage**

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35 **ABSTRACT**

36 *Carbon capture and utilization technologies, along with the use of lightweight concrete, offer*  
37 *promising solutions towards the decarbonization of cement industry. Combining both practices, this*  
38 *paper proposes an innovative type of foamed concrete produced with CO<sub>2</sub> gas foam instead of*  
39 *common air foam. The objective is to demonstrate the viability of inducing the formation of porosity*  
40 *in the material, which is necessary for its optimal structure, while simultaneously enabling the*  
41 *permanent storage of carbon dioxide through the chemical reaction between the CO<sub>2</sub> gas and the*  
42 *fresh cementitious paste. Samples are produced using the preformed foaming method, injecting CO<sub>2</sub>*  
43 *gas instead of air, and mixing foam and cement paste prior to casting in prismatic moulds. Common*  
44 *foamed concrete samples produced with air foam are considered as reference.*  
45 *Compressive tests are carried out to investigate the mechanical properties. X-Ray Diffractometry and*  
46 *Scanning Electron Microscopy are conducted to investigate influence of CO<sub>2</sub> gas on the*  
47 *microstructure of the foamed concrete. Thermogravimetry is adopted to quantify the stored CO<sub>2</sub>.*  
48 *The results obtained from this study are extremely encouraging. The use of CO<sub>2</sub> foam was found to*  
49 *nearly double the CO<sub>2</sub> uptake, without producing any significant change in the compressive strength-*  
50 *to-density ratio. Furthermore, the utilisation of CO<sub>2</sub> foam demonstrated to exert a favourable*  
51 *influence on the microstructure of the material, resulting in the formation of smaller pores that exhibit*  
52 *a more uniform size distribution.*

53

54 **KEYWORDS:** CO<sub>2</sub> Foamed Concrete; Compressive strength; Pore size distribution; CO<sub>2</sub>  
55 mineralisation.

56

## 57 **1.INTRODUCTION**

58 Cement is the principal material adopted in the construction field, due to the global increase in  
59 demand for concrete (Mohamad et al. 2022). Consequently, the cement industry is responsible for  
60 approximately 7% of global CO<sub>2</sub> emissions (Korczak et al. 2022). In order to comply with the EU  
61 climate policy standards, the sector is required to reduce CO<sub>2</sub> emissions by 30% by the year 2030 and  
62 to achieve carbon neutrality by the year 2050 (Cembureau 2021). In this regard, strategies for Carbon  
63 Capture Storage and Utilization (CCS/CCU) account for 36% of the overall reduction. CCS actions  
64 consist of the reduction of greenhouse gas emissions through CO<sub>2</sub> mineralisation processes. CCU is  
65 used to denote processes in which carbon dioxide is stored and the products are valuable for a practical  
66 use.

67 Several CO<sub>2</sub> utilisation technologies for construction materials have emerged. These processes  
68 involve the reuse of captured CO<sub>2</sub> in the curing, production and modification of building materials  
69 (Li et al. 2022). The majority of employed mineralisation techniques include: the use of a carbonation  
70 chamber for the curing of pre-cast elements (Ferrara et al. 2021; Librandi et al. 2019); accelerated  
71 aqueous mineralisation processes (Bonfante<sup>1</sup> et al. 2024; Bonfante<sup>2</sup> et al. 2024) to produce carbonated  
72 powders that are adopted as fillers or cement replacements (Bonfante et al. 2025; Fang et al. 2022).  
73 Recently, CO<sub>2</sub> injection during the mixing process to produce fresh concrete, has emerged as  
74 alternative CCU strategy. This novel approach represents a significant departure from the well-  
75 established mineralisation processes mentioned above (Ferrara et al. 2024). The formation of calcium  
76 carbonate during the fresh mixture resulted in an acceleration of the hydration reactions (Davolio et  
77 al. 2025). A limited number of pioneering studies have employed a comparable methodology in order  
78 to investigate the potential of utilising CO<sub>2</sub> gas in fresh pastes to develop foamed concrete. In this  
79 process, gaseous CO<sub>2</sub> is employed for the generation of bubbles and for reacting with the paste,  
80 resulting in the formation of stable calcium carbonates (Ta<sup>1</sup> et al. 2023; Ta<sup>2</sup> et al. 2023).

81 Foamed concrete is a specialised form of lightweight concrete, in which the reduction in weight is  
82 achieved through the incorporation of air pores within the concrete mixture. The latter can be  
83 introduced by either the mix-foaming method or the pre-foaming method (Amran et al. 2020). In the  
84 former case, the liquid foaming agent is introduced directly alongside with the other ingredients in  
85 the mix design, and the cellular system is generated during the turbulent and vigorous mixing phase.  
86 In the second case, the foaming agent is employed to generate the preformed foam, which is  
87 subsequently incorporated into the cementitious conglomerate and mixed until a homogeneous  
88 system is attained.

89 The increasing interest in foamed concrete can be attributed to the straightforward manipulation of  
90 porosity, which confers a high degree of versatility in its applications. Indeed, low and medium-low  
91 density specimens have been shown to be particularly suited to applications requiring thermal  
92 insulation and fire resistance properties. As the density of the material increases, the potential for  
93 exploitation as structural materials also grows. This is particularly advantageous in areas of high  
94 seismicity (Falliano et al. 2023). Furthermore, foamed concrete is typically distinguished by a high  
95 thermal inertia with a time lag comparable to that of conventional materials, yet with a higher  
96 decrement, rendering it suitable also for applications in warm environments (Ropelwski et al. 1999).  
97 In recent years, there has been a growing interest in the feasibility of utilising CO<sub>2</sub> as a foaming agent  
98 for the production of lightweight foamed concrete attracted interest. In their study, Li et al. 2020,  
99 examined the thermal insulation performance of foamed concrete produced with varying foaming  
100 gases. The findings of the study indicate that the lower thermal conductivity of CO<sub>2</sub> gas compared to  
101 air resulted in the CO<sub>2</sub> foamed concrete exhibiting enhanced thermal insulation compared to  
102 traditional foamed concrete. Whilst the objective of the study was not to induce CO<sub>2</sub> mineralisation  
103 for carbon storage, it demonstrated the feasibility of producing CO<sub>2</sub> foam.

104 A number of studies have proposed the preparation of CO<sub>2</sub> foamed concrete by means of a chemical  
105 foaming method. This process entails the utilisation of compounds that undergo a reaction with the  
106 cementitious paste to produce CO<sub>2</sub> gas, acting as foaming agents (Yao et al. 2022). Examples of this

107 include the utilisation of rapid hardening cement in the paste, as well as the incorporation of sodium  
108 bicarbonate or aluminium salt as a foaming agent (Ma et al. 2017). Nevertheless, the physical foaming  
109 method, which involves the generation of foam directly from a gas, was identified as a more efficient  
110 technique for controlling bubble size and distribution (Phavongkham et al. 2020).

111 To date, only a limited number of studies have explored the potential for creating foamed concrete  
112 using carbon dioxide gas (physical foaming method), with a particular focus on the effects of both  
113 mechanical behaviour and carbon storage (Ta<sup>1</sup> et al. 2023; Ta<sup>2</sup> et al. 2023; Liu et al. 2023). In their  
114 study, Liu et al. 2023 produced CO<sub>2</sub> modified foam concrete, which exhibiting a notable enhancement  
115 in strength compared to the reference sample. Furthermore, the presence of CO<sub>2</sub> gas within the pores  
116 was observed to facilitate a greater formation of calcium carbonate. Nevertheless, the assessment of  
117 carbon storage was confined to qualitative examinations, lacking the provision of data pertaining to  
118 CO<sub>2</sub> uptake. The results obtained were encouraging, with lightened samples having a dry density of  
119 597 kg/m<sup>3</sup>, CO<sub>2</sub> uptake of 8.57% and compressive strength of 1.75 MPa (Ta<sup>1</sup> et al. 2023). However,  
120 a carbonation pre-treatment was required prior to the addition of CO<sub>2</sub> foam to increase the viscosity  
121 of the paste. This process involved the mixing the cement slurry with CO<sub>2</sub> gas for a duration of up to  
122 120 min, with consequent acceleration in the hydration process (Zajac et al. 2020) that may have  
123 compromised the development of strength during the curing period. In the study conducted by Zhang  
124 et al. 2024, foamed concrete was produced by partially substituting the conventional foam stabiliser  
125 (polyacrylic amide) with CO<sub>2</sub>. This substitution resulted in enhanced strength when compared to the  
126 control samples.

127 A critical issue associated with the use of carbon dioxide to generate the foam is the release of CO<sub>2</sub>  
128 gas into the atmosphere during the process. In this regard, Ta<sup>2</sup> et al. 2023 emphasised the importance  
129 of enhancing the stability of the CO<sub>2</sub> foam in order to reduce the gas release. Furthermore, the  
130 utilisation of solid waste materials, such as coal gangue and fly ash, is proposed to enhance carbon  
131 sequestration capacity, thereby reducing the environmental impact of the process.

132 The poor stability of CO<sub>2</sub> foam, and its reactivity with portlandite cement, pose significant challenges  
133 to the production of CO<sub>2</sub> foamed concrete. The establishment of a clear procedure for the creation of  
134 an efficient material is not yet established. The controversial outcomes pertaining to strength, in  
135 conjunction with the absence of a precise quantification of the carbonation potential, underscore the  
136 necessity for additional investigation to enhance the performance of this innovative material and to  
137 delineate its mechanical and environmental capabilities.

138 In view of the aforementioned considerations, the aim of the present study is to provide a  
139 comprehensive evaluation of the potential for CO<sub>2</sub> foamed concrete. In order to achieve this objective,  
140 the investigation encompasses the mechanical properties and microstructure of the material, in  
141 addition to the evaluation of the stored carbon to define the impact of the process as a CCU technique.  
142 The generation of the foam is achieved through the mixing of CO<sub>2</sub> gas with a surfactant, which is  
143 then added to the cementitious paste in order to create the foamed concrete. Any pre-treatment is  
144 employed to produce cement paste. Results are compared with similar studies from the literature.  
145 Notably, no additional substances were introduced to improve the stability of the CO<sub>2</sub> foam, ensuring  
146 that the comparison focuses solely on the impact of the foaming gas.

## 147 2.RAW MATERIALS

148 The binder used in the study is Portland cement, and specifically CEM 52.5 R type. The particle size  
149 distribution, determined by laser granulometry (Malvern Mastersizer 3000E laser diffraction particle  
150 size analyser), exhibited characteristic values d10, d50 and d90 of 1.87  $\mu\text{m}$ , 11.3  $\mu\text{m}$  and 30.6  $\mu\text{m}$   
151 respectively (see Figure 1).

152 The elemental composition, determined by X-ray fluorescence analysis (XRF, Rigaku Supermini  
153 2000), is presented in Table 1.

154 The mineralogical composition was investigated by X-ray diffraction analysis (XRD, Empyrean  
155 Malvern Panalytical) in the Bragg-Brentano configuration, with Cu-K $\alpha$  wavelength radiation  
156 (0.15406 nm) operating at 40 kV and 40 mA. The  $2\theta$  range was from  $5^\circ$  to  $70^\circ$  with an angular step  
157 of  $0.006^\circ$ , and this was held for 23 s. The diffractometer was equipped with Soller ( $0.04$  rad), anti-  
158 scatter (P7.5), beam mask (10 mm), divergence ( $\frac{1}{4}^\circ$ ) and axial ( $\frac{1}{2}^\circ$ ) slits. The XRD spectra are  
159 displayed in Figure 2. As expected for cement, the main compounds identified were calcium silicates  
160 (belite and alite), aluminoferrites, aluminates and calcium sulphates. The presence of calcium  
161 carbonate was also noted.

162 Thermogravimetric differential thermal analysis (TG-DTA, LabSys evo machine Setaram) was  
163 carried out in an  $\text{N}_2$  atmosphere at a heating rate of  $10^\circ\text{C}/\text{min}$  up to  $1050^\circ\text{C}$  (Figure 3). The curves  
164 illustrate the dehydration of gypsum at approximately  $140^\circ\text{C}$  (peak I), the dehydration of portlandite  
165 at around  $450^\circ\text{C}$  (peak II), the decomposition of calcium carbonate at approximately  $750^\circ\text{C}$  (peak  
166 III). In accordance with the XRD pattern, the minimal mass loss (0.2%) concomitant with portlandite  
167 indicates a negligible presence of this compound. The initial  $\text{CO}_2$  content,  $\%\text{CO}_{2,\text{cem}}$ , was estimated  
168 to be 2.3%.

169 The theoretical maximum  $\text{CO}_2$  uptake, as determined by Steinour equation (Huntzinger et al. 2009),  
170 was calculated to be 47.8%, based on the XRF elemental composition. This value, which represents  
171 the carbonation potential that would theoretically be reached if all the available calcium reacted with

172 CO<sub>2</sub>, was utilised to ascertain the carbonation efficiency,  $\eta_{\text{carb}}$ , defined as the ratio between the  
173 experimental CO<sub>2,uptake</sub> and the theoretical one.

174 In this study, the creation of air voids was achieved through the utilisation of a protein foaming agent,  
175 Isocem, produced by Isoltech srl. As has been previously emphasised in the relevant literature, the  
176 use of a protein-based foaming agent results in superior mechanical properties in the final foamed  
177 concrete in comparison to synthetic foames (Falliano et al. 2018; Falliano et al. 2021). The dark brown  
178 liquid foaming agent was characterised by a specific weight of  $1.15\pm 0.2$  g/ml, and by a pH of 6.75.  
179 Furthermore, the experimental campaign involved tap water, and MasterEase®, a polycarboxylate  
180 superplasticiser produced by Master Builders.

## 181 **3.METHODS**

### 182 **3.1 Definition of operational parameters**

183 The study encompasses two series of samples, comprising air and CO<sub>2</sub> foam, respectively. The  
184 operational parameters required to produce both the foamed concrete were accurately defined, based  
185 on previous studies conducted by the authors. The key factors that must be considered are the pressure  
186 of the gas, the concentration of the foaming agent, the mixing rotational speed, water-to-cement ratio,  
187 superplasticiser-to-cement ratio, fresh concrete density. This section is dedicated to the presentation  
188 of the rationale behind the selection of the parameters.

189 The production of foam, employing both air and CO<sub>2</sub>, was conducted utilising the same foam  
190 generator, Figure 4. The operational mechanics of the apparatus are analogous to those exhaustively  
191 delineated in Falliano et al. (2021). It has been demonstrated that the drainage of the foam produced  
192 is significantly influenced by pressure and the concentration of the foaming agent. Drainage, which  
193 is closely associated with the lifetime of the foam and, consequently, to the quality of the foamed  
194 concrete produced, is markedly reduced when the pressure used is above 2.5 bar and when the  
195 foaming agent concentration is above 3% (Falliano et al. 2021). The pressure employed for foam  
196 generation was approximately 3 bar, while the surfactant concentration was selected to be 5%.

197 As demonstrated by Falliano et al. (2020), superior performance – both physical and mechanical – is  
198 facilitated by the utilisation of elevated mixing intensities, thereby enhancing the foamed concrete  
199 microstructure. The mixing process was conducted using a vertical mixer at a rotational speed of 3000  
200 rpm.

201 In both the cases of air and CO<sub>2</sub> foam production, an identical mix proportion was adopted in the  
202 creation of the foamed concrete presented in this study. In previous studies (Falliano et al., 2018) and  
203 (Falliano et al., 2022), it was established that fresh foamed concrete demonstrated stability across a  
204 wide range of solid-to-liquid ratios (i.e., foam + water-to-cement ratio). In consideration of the

205 aforementioned parameters, the water-to-cement ratio was determined to be 0.33, and the  
206 superplasticizer-to-cement ratio was established at 0.15.

207 In light of the higher foam density produced using CO<sub>2</sub> in comparison to that generated using air, a  
208 different foam-to-cement ratio was necessitated in each case to achieve the same target density.  
209 Specifically, the quantity of CO<sub>2</sub> foam adopted in order to achieve the target fresh density of 750±50  
210 kg/m<sup>3</sup> was precisely 6.5 times greater than the air foam necessary to obtain the same density. In  
211 addition to density, liquid drainage, closely linked to foam lifetime (Falliano et al, 2021), proved to  
212 be a key distinction between air-generated foam and CO<sub>2</sub>-generated foam. Specifically, while air-  
213 generated foam exhibited no detectable drainage even ten minutes after preparation, CO<sub>2</sub>-generated  
214 foam showed measurable drainage within two minutes of its production. The use of CO<sub>2</sub> instead of  
215 air results in greater liquid drainage and, consequently, increased foam instability. Indeed, compared  
216 to the use of air, CO<sub>2</sub> promotes surface tension-driven drainage and gas diffusion. CO<sub>2</sub> is significantly  
217 more soluble in water than air. Utilizing a gas with very low solubility in water slows down gas  
218 transport across the aqueous phase compared to a more soluble gas (Fameau et Salonen, 2014). This  
219 further explains the greater instability observed in CO<sub>2</sub>-based foams compared to those produced with  
220 air. Furthermore, the higher instability of CO<sub>2</sub>-based foam is also attributed to a greater degree of  
221 bubble coalescence, a phenomenon triggered by the higher density of CO<sub>2</sub> compared to air (Liu et al,  
222 2023). This coalescence refers to the physical process by which liquid droplets, gas bubbles, or solid  
223 particles merge to form a single larger entity. For this reason, the foam was immediately mixed with  
224 the cementitious paste after its generation to prevent the drainage phenomenon. This is a crucial  
225 approach for ensuring the production of high-quality foamed concrete.

### 226 **3.2 Sample preparation and mechanical testing procedures**

227 Foamed concrete samples produced with air foam are designated as FC\_Air, while foamed concrete  
228 samples produced with CO<sub>2</sub> foam are designated as FC\_CO<sub>2</sub>. The foam generator was connected to  
229 an air compressor in order to generate air foam. In the case of the FC\_CO<sub>2</sub> specimens, the generator

230 was connected to a CO<sub>2</sub> cylinder with gas purity of 99.9% (grade 2.5). Initially, the cement, water,  
231 and superplasticiser were mixed until a homogeneous cementitious system was obtained.  
232 Immediately following its generation, the requisite quantity of foam was added to the previously  
233 prepared cementitious mixture to attain the target fresh density, i.e. 750±50 kg/m<sup>3</sup>. The mixing  
234 process was continued for a further two minutes or until a homogeneous system was achieved, in any  
235 case. Subsequently, the material was accurately poured in order to guarantee optimal adherence to  
236 the formwork. Following a period of 48 hours, the prismatic samples, with dimensions of 20 mm x  
237 20 mm x 80 mm, were demoulded and cured in air at room temperature (20±3°C) and relative  
238 humidity of 65±5%. The sample size adopted is commonly used in the literature for the  
239 characterization of cementitious pastes (Suarez-Riera et al. 2020). For each series, four samples were  
240 cast. Three of them were subjected to mechanical testing, while a fourth sample was used for chemical  
241 characterisation. Following the curing process, the specimens were subjected to three-point bending  
242 tests and compressive tests. These tests were conducted in accordance with the procedure outlined in  
243 EN 196-1. The testing procedure involved the application of half of the specified loading rate,  
244 calculated based on the scaled sample size. The flexural strength evaluation was carried out in force-  
245 controlled mode through a Zwick-Line Z050 testing machine, with a loading rate of 25 N/s, applying  
246 the load perpendicularly to the direction of casting. The span length was equal to 60 mm.  
247 The compressive strength was evaluated in force control mode, utilising the Zwick-Line testing  
248 apparatus, in addition to the halves of the prisms derived from the three-point bending test. The  
249 specimens were subjected to a loading rate of 1200 N/s, applied perpendicularly to the direction of  
250 casting. Following the testing phase, the specimens were placed in an oven and dried at a temperature  
251 of 100°C, in accordance with standard practice for assessing the density of hardened mixes of  
252 cementitious conglomerates. This process was continued for a minimum of 48 hours or until the  
253 specimens reached a constant weight, in order to evaluate their dry density. Figure 5 reports a scheme  
254 of the process adopted to produce and characterise the foamed samples.

### 255 3.3 Carbon capture assessment

256 This section delineates the methodologies employed for the estimation of the CO<sub>2</sub> chemically bound  
257 within the material as a consequence of the formation of calcium carbonate. The efficiency of the  
258 carbonation process was evaluated through the analysis of the hardened foamed paste specimens. In  
259 particular, a sample was extracted from the core of prismatic specimens 24 hours after casting, milled,  
260 and immediately subjected to the analysis. The sample was not subjected to a drying process in order  
261 to prevent the additional carbonation of portlandite and the hydration of calcium silicates present in  
262 the cement paste, which could occur as a result of prolonged exposure to the atmosphere. XRD and  
263 SEM analysis were adopted to qualitatively investigate the formation of calcium carbonate. TG-DTA  
264 analysis was adopted to quantify the calcium carbonate content, which is related to the mass loss in  
265 the temperature range of 550°C to 850°C (Ferrara et al. 2023). The CO<sub>2</sub> content is expressed as A  
266 percentage of the mass with respect to the amount of binder. The latter is considered to be equal to  
267 the TG value at a temperature of 950 °C (Mo et al. 2016), after the release of water and CO<sub>2</sub> content  
268 deriving from hydration and carbonation reactions (Equation 1):

$$\%CO_2 = \frac{\% \Delta W_{550^{\circ}C-850^{\circ}C}}{\%W_{950^{\circ}C}} \quad \text{Equation 1}$$

269 where  $\% \Delta W_{550^{\circ}C-850^{\circ}C}$  denotes the %TG mass loss in the 550°C-850°C range of temperature, and  
270  $\%W_{950^{\circ}C}$  denotes the %TG at 950°C.  $\%CO_{2,uptake}$  is then defined as the difference between the %CO<sub>2</sub>  
271 and the initial CO<sub>2</sub> content of the raw cement powder, designated as  $\%CO_{2, cem}$ .

272 The portlandite content in the hardened samples, %Ca(OH)<sub>2</sub>, is determined from TG-DTA curves  
273 that exhibit a mass loss due to the release of water during the dihydroxylation calcium hydroxide. In  
274 order to eliminate the contribution of C-S-H decomposition from the estimate, the tangent method  
275 was adopted (Lothenbach et al. 2016). In addition to the CO<sub>2</sub> content, the portlandite content is also  
276 expressed as percentage of the mass at 950 °C.

277 The TG curve at temperatures below 150 °C was characterised by a significant mass loss, which was  
278 attributed to the release of humidity and the physical water bond with hydrate calcium silicates. In

279 order to facilitate a more accurate representation of mass losses at elevated temperatures, which are  
280 influenced by the presence of portlandite and carbonates, the initial significant mass loss was  
281 excluded from the analysis. Conversely, the TG values were plotted assuming a temperature of 150°C,  
282 which was considered as equivalent to 100%.

283 Furthermore, the XRD analysis was conducted to examine the phase transformation of the various  
284 compounds throughout the process and to corroborate the quantitative analysis conducted via TG-  
285 DTA. The adopted procedures are outlined in section 2. Field-emission Scanning Electron  
286 Microscope (SEM) images were generated at different magnifications to investigate the  
287 microstructure of the hardened samples. The image manipulation software GIMP was adopted for the  
288 quantification of the average size of the pores within the hardened material. The data were collected  
289 with the objective of defining the pore size cumulative distribution as demonstrated by existing  
290 literature focused on the analysis of the microstructure of foamed concrete (Hilal et al. 2015).

## 291 4.RESULTS AND DISCUSSION

### 292 4.1 Mineral composition and carbon capture

293 Figure 6 reports the XRD pattern of FC\_Air and FC\_CO<sub>2</sub> samples, indicating the main crystalline  
294 compounds present in the material. XRD pattern of raw cement is also reported for comparison. As  
295 expected, the FC\_Air sample displayed the presence of portlandite, a product of calcium silicates  
296 hydration, and ettringite, a product of the reaction between gypsum and calcium aluminates. The  
297 presence of calcium silicates hydrates, C-S-H, presumed to result from the hydration of belite and  
298 alite, was not discernible due to their low crystallinity. Furthermore, peaks attributable to the primary  
299 compounds of raw cement, in addition to a number of calcite peaks, were also discerned.

300 The XRD pattern of the FC\_CO<sub>2</sub> sample exhibited analogous compounds to those identified in the  
301 FC\_Air sample. However, an alteration in the relative intensity of the peaks was observed with an  
302 increase in the signal related to calcite in comparison to portlandite. This may indicate, at least  
303 qualitatively, that a portion of the portlandite derived from the hydration process reacted with the CO<sub>2</sub>  
304 present in the foam, forming calcium carbonates. Furthermore, the XRD pattern exhibited peaks  
305 associated with calcium hemicarboaluminate, displaying heightened intensity in comparison to the  
306 FC\_Air sample. This compound is typically present in the early stages of hydration in Portland  
307 cement with calcite content, resulting from the reaction of calcium aluminates. As demonstrated by  
308 Ipavec et al. (2011), the substance undergoes a transformation into a more stable monocarboaluminate  
309 during periods of prolonged hydration. Consequently, the higher presence in FC\_CO<sub>2</sub> samples also  
310 corroborates the occurrence of a carbonation process. The presence of mono- and  
311 hemicarboaluminates exerts a stabilising effect on ettringite (Zajac et al. 2014). The consequent  
312 reduction in the availability of hydrated calcium aluminates (C-A-H) limits its capacity to react with  
313 gypsum from sulphatic attack, thus preventing the formation of secondary ettringite.

314 Figure 7 and Figure 8, respectively, illustrate the TG-DTA curves of the FC\_Air and FC\_CO<sub>2</sub>  
315 samples. The peak observed in DTA curve at approximately 250 °C, peak I, is associated with the

316 decomposition of AFm phases, which may be either sulfate or carbonate, derived from the hydration  
317 of aluminates (Hadj Sadok et al. 2021; Alzeer et al. 2022). Peak II, situated at approximately 470°C,  
318 corresponds to the dehydration of portlandite, which induces the considerable mass loss observed in  
319 the TG curve. Peak III is associated with the decomposition of calcium carbonate, which may exist  
320 in various crystalline forms, as evidenced by the presence of preceding minor peak. The absence of  
321 this minor peak in the raw material indicates that it is a consequence of the manufacturing process in  
322 the presence of CO<sub>2</sub>.

323 Table 2 presents the data regarding the CO<sub>2</sub> content resulting from the decomposition of calcium  
324 carbonate, and the water content resulting from the dihydroxylation of portlandite. Additionally. It  
325 includes the calcite and portlandite contents for both samples.

326 The findings of the quantitative analysis demonstrated that the sample with a higher carbonation  
327 degree exhibited a lower portlandite content, thereby corroborating the observation made through  
328 XRD analysis. In greater detail, the presence of CO<sub>2</sub> foam in the FC\_CO<sub>2</sub> sample resulted in a  
329 reduction in portlandite content, %Ca(OH)<sub>2</sub>, by 3.6% in comparison to the FC\_Air sample (as  
330 evidenced by the data in Table 1). Assuming that the formation of one mole of calcium carbonate is  
331 derived from the mineralisation of each mole of portlandite, an increase in the carbon dioxide content,  
332 %CO<sub>2</sub>, of 2.1% should be indicative of this variation in portlandite content. However, the results in  
333 Table 1 indicate a higher %CO<sub>2</sub> increase, corresponding to 3.9%. The observed phenomenon can be  
334 attributed to the varying reactivity of calcium silicates in the presence of CO<sub>2</sub>. Indeed, the processes  
335 of hydration and carbonation in cement are analogous, characterised by dissolution and precipitation  
336 reactions. Nevertheless, while the higher degree of hydration of calcium silicates is typically achieved  
337 within several hours, it is demonstrated to be achieved after only 30 minutes during accelerated  
338 carbonation (Zajac et al. 2020). It is therefore reasonable to hypostasise that the CO<sub>2</sub> present in the  
339 pores of the FC\_CO<sub>2</sub> sample during the casting process, and dissolved in the mixture, accelerates the  
340 hydration process. Consequently, a greater quantity of hydration products is produced, which are then  
341 available for carbonation.

342 The  $\text{CO}_{2,\text{uptake}}$  of the FC\_Air sample was found to be 4.7%, which can be attributed to natural  
343 carbonation in atmospheric conditions. This value is consistent with the findings of previous studies,  
344 which reported a value of 4.14% for a plain aerated sample (Zhang et al. 2024). The  $\text{CO}_{2,\text{uptake}}$  of the  
345 FC\_  $\text{CO}_2$  sample was found to be equal to 8.7%. This outcome is of particular interest when considered  
346 in the context of analogous research conducted on  $\text{CO}_2$  foamed concrete, which has documented  
347  $\text{CO}_2$  uptake values ranging from 6.10% to 8.57% (Ta<sup>1</sup> et al. 2023; Ta<sup>2</sup> et al. 2023) and from 4.20% to  
348 7.48% (Zhang et al. 2024).

349 The carbonation efficiency,  $\eta_{\text{carb}}$ , was found to be equal to 9.9% and 18.1% for the FC\_Air and  
350 FC\_  $\text{CO}_2$  samples, respectively. It was observed that the utilisation of  $\text{CO}_2$  foam effectively induced  
351 a mineralisation process in the fresh cement paste, resulting in a doubling of the amount of carbon  
352 dioxide with respect to the foamed concrete cast with conventional air foam.

353 It is noteworthy that the  $\text{CO}_2$  uptake parameter is used to describe the capacity of the material to  
354 permanently store  $\text{CO}_2$  as a consequence of the formation of calcium carbonate. This parameter is of  
355 great importance in the definition of the overall carbon footprint of the foamed concrete production  
356 process. However, a more comprehensive analysis is required in order to effectively define the net  
357  $\text{CO}_2$  balance of the process. A primary and crucial aspect is the release of  $\text{CO}_2$  into the atmosphere  
358 during the foaming generation process. In this regard, enhancing the stability of the foam can mitigate  
359 the dispersion of the gas. Furthermore, it would reduce the duration of the generation process, which  
360 is already of the order of a few minutes, thereby further reducing this dispersion. In addition, as  
361 proposed in the relevant literature (Ta<sup>1</sup> et al. 2023), an eco-efficient process should be integrated with  
362 other industrial plants that provide flue gas for use as a  $\text{CO}_2$  source, and wastes to be used in  
363 combination with cement. The circulation of both gas and by-products establishes a virtuous cycle  
364 that serves to reduce the overall impact of the production system.

## 365 4.2 Mechanical strength

366 Figure 9 presents a comparison of the dry densities of the two distinct types of foamed concrete under  
367 investigation. Indeed, in order to make an effective comparison of the mechanical properties of  
368 foamed concrete, it is essential to evaluate its dry density. This methodological approach facilitates  
369 more precise comparisons with other studies in the scientific literature, where foamed concrete is  
370 characterised by analogous dry densities, thus providing a consistent basis for analysing performance  
371 across disparate investigations. The findings of the experiments conducted in order to ascertain the  
372 compressive and flexural strengths are presented in Figure 10. Moreover, all the figures present the  
373 mean values with standard deviations, alongside the experimental data, in order to provide a  
374 quantitative assessment of the dispersion of the findings. While the fresh density is approximately  
375 equivalent, the latter figure demonstrates that the discrepancy in terms of dry density is considerably  
376 more pronounced. Indeed, samples prepared with CO<sub>2</sub> foam exhibit a reduction in dry density of  
377 approximately 15%, which is slightly less than that observed in samples with traditional foam. This  
378 is an anticipated outcome, as evidenced by the higher quantity of CO<sub>2</sub> foam, as illustrated in Section  
379 3.1, necessary to attain the target fresh density of 750±50 kg/m<sup>3</sup>. It was expected that more CO<sub>2</sub> foam  
380 would be needed to achieve the desired target fresh density, since CO<sub>2</sub> foam produced was about 6.5  
381 times denser than air foam. This increased the liquid-to-solid ratio, as reflected in the lower dry  
382 density of CO<sub>2</sub>-based foamed concrete. However, no coalescence or instability was observed, which  
383 can be attributed to the immediate mixing of the foam with the cementitious paste. This stability is  
384 attributed to the very low water-to-cement ratio and the carefully designed mix proportions, based on  
385 prior expertise (Falliano et al., 2018; Falliano et al., 2022), which ensured stability across a wide  
386 liquid-to-solid ratio range, including those used in this study. Despite the non-negligible reduction in  
387 dry density, which is contrary to the general expectation (Nambiar et Ramamurthy 2007), the CO<sub>2</sub>  
388 foam specimens exhibited a microstructure with voids characterised by smaller diameters and a more  
389 homogeneous distribution. This aspect will be outlined in greater detail in Section 4.3. This significant  
390 finding enables the ratio of compressive strength, MPa, to dry density, kg/m<sup>3</sup>, ( $\sigma_c/\rho$ ) to be maintained

391 roughly constant. This ratio typically declines as the material approaches the threshold of  
392 ultralightweight foamed concretes. For instance, ultralightweight foamed concretes, defined as  
393 having a dry density below  $100 \text{ kg/m}^3$ , exhibit a ratio of compressive strength to dry density ( $\sigma_c/\rho$ )  
394 ranging from 0.0017 to 0.0029 (Falliano et al. 2022). In this study, both for the air and the  $\text{CO}_2$  foamed  
395 concretes presented in this study,  $\sigma_c/\rho$  is approximately the same, with values of 0.0076, and 0.0077,  
396 respectively.

397 It is established that the presence of bubbles of a smaller size and a more homogeneous distribution  
398 in foamed concretes improves performance (Falliano et al. 2019). This noteworthy outcome can be  
399 further elucidated by comparing it with the compressive strengths of other foamed concretes with  
400 similar densities, which have already been discussed in the relevant literature, but obtained from air  
401 foam. For example, in (Koksal et al. 2020), foamed concretes with a density of approximately  $600$   
402  $\text{kg/m}^3$  are characterised by a ratio of compressive strength, MPa, to dry density,  $\text{kg/m}^3$ , approximately  
403 equal to 0.006; moving towards  $500 \text{ kg/m}^3$ , in (Liu et al. 2023) this ratio is equal to 0.0066, in (Hou  
404 et al. 2019) is even lower, and approximately equal to 0.006. It must be highlighted that the strength  
405 to dry density ratio was used for comparison as representative of the overall performance of the  
406 lightweight foamed concrete. However, due to a lack of studies concerning  $\text{CO}_2$  foamed concrete, the  
407 compared data differ in composition and manufacturing process. These aspects must be considered  
408 to fully understand the different behaviours. For example, in Koksal et al. (2020), expanded  
409 vermiculite powder was adopted as aggregate in the foam concrete mixture, while no aggregates were  
410 used in the present study. Moreover, they included fly ash in the cement blend. In Liu et al. (2023),  
411 hydroxypropyl methyl cellulose (HPMC) was employed for foam stabilization and water retention.  
412 In Hou et al. (2019), nanoparticles such as graphene and hydrophilic nano- $\text{SiO}_2$  were added to the  
413 foaming agent during the foam generation. Nevertheless, these comparisons demonstrate the  
414 exceptional compressive strength results achieved by the  $\text{CO}_2$  foamed concrete presented in this  
415 study. These excellent results are further substantiated when a comparison is made with the  $\text{CO}_2$   
416 foamed concrete presented in the study from Ta<sup>1</sup> et al. 2023, Portland cement was subjected to

417 carbonation pre-treatment, in contrast to the methodology employed in this study. In fact, as  
418 demonstrated in (Ta<sup>1</sup> et al. 2023), foamed concretes with densities of 597 kg/m<sup>3</sup> are characterised by  
419 a ratio of compressive strength, MPa, to dry density, kg/m<sup>3</sup>, equal to 0.0029, while in the case of  
420 densities of 621 kg/m<sup>3</sup> and 669 kg/m<sup>3</sup>, this ratio increases to 0.003 and 0.0036, respectively.  
421 Moreover, in Liu et al. (2023), for samples made with 100% CO<sub>2</sub> foam, the compressive strength-to-  
422 density ratio is approximately 0.0045. The enhanced outcomes observed in the present study are  
423 presumably attributable to the different procedure employed in the preparation of CO<sub>2</sub> foam, which  
424 facilitated the generation of a stable CO<sub>2</sub> foam with sufficient duration to be mixed with the cement  
425 paste without the incorporation of HPMC. In contrast, Liu et al. (2023) generated the foam by stirring  
426 for 150 seconds at a speed of 1800 r/min in a confined space containing CO<sub>2</sub>, introducing the gas into  
427 the foam from the bottom. In this study, as mentioned above, a proper foam generator (Falliano et al.,  
428 2020) was employed. Therefore, despite the very close densities, the ratio previously shown is  
429 significantly lower than that characterising the CO<sub>2</sub> foamed concrete presented in this paper. The  
430 comparison with the literature of the results in terms of strength to dry density ratio is reported in  
431 Figure 11.

432 With regard to flexural strength, CO<sub>2</sub> foamed concrete demonstrated a lower performance, a  
433 consequence not only of its reduced density compared to air foamed concrete, but also in comparison  
434 with the bending strength-to-density ratio ( $\sigma_f/\rho$ ). The latter ratio was found to be equal to 0.0027 and  
435 0.0018 for air and CO<sub>2</sub> foamed concrete, respectively. A subsequent study will provide a more  
436 comprehensive analysis of this behaviour. It is imperative to emphasise that, considering the potential  
437 applications of this material in non-structural contexts, the most significant mechanical performances  
438 are undoubtedly those related to its compressive strength.

### 439 **4.3 Microstructure**

440 Figure 12 depicts SEM images of the hardened FC\_Air and FC\_CO<sub>2</sub> samples at varying  
441 magnifications.

442 Representative sections of both the FC\_Air and FC\_CO<sub>2</sub> samples are illustrated in Figure 12a and b,  
443 at the lowest magnification, and show the pores generated by the foam during the mixing process that  
444 remained in the hardened state. The distribution of pores sizes generated by the foam was subjected  
445 to statistical analysis for both samples. A representative continuous region, comprising a population  
446 of approximately 2000 pores, was selected for analysis in both the FC\_Air and FC\_CO<sub>2</sub> samples. The  
447 diameter of each pore was then measured. The pores were observed to have a relatively circular shape.  
448 In the case of irregular pores, the shortest diameter was deemed to be the most appropriate for  
449 consideration. The mean values of pore size were found to be 59.3 μm (St.Dev. 53.7μm) and 52.0μm  
450 (St.Dev. 37.7μm), respectively, for the FC\_Air and FC\_CO<sub>2</sub> samples. To gain further insight into the  
451 pore size distribution and to better comprehend the observed variability, the results from the two  
452 samples were compared in terms of the area occupied by the pores within the image region of analysis.  
453 In order to achieve this objective, the assumption was made that the SEM images were representative  
454 of a plane section, and the area of each pore was calculated from the diameter data, assuming a circular  
455 shape. The results are presented in Figure 13, where the pore size is reported on the x-axis and the  
456 cumulative distribution in terms of area is expressed as a percentage of the total area occupied by the  
457 considered pore population on the y-axis.

458 The curves demonstrate an upper section characterised by irregular, minor steps, indicating a  
459 discontinuous distribution due to the presence of pores with markedly larger dimensions. This is  
460 followed by a continuous distribution indicating less pronounced difference in size between  
461 consecutive data points. The chart demonstrates that the FC\_CO<sub>2</sub> sample exhibited a distribution of  
462 smaller pores in comparison to the FC\_Air sample across the entire population. Moreover, the  
463 discontinuous part in the FC\_Air curve, which is more pronounced than in the FC\_CO<sub>2</sub> curve,  
464 indicates that approximately 30% of the total area is occupied by pores with a size much larger than  
465 the average. The statistical analysis confirms the trend that can be seen visually in Figure 12 a and b.  
466 In fact, the use of CO<sub>2</sub> foam resulted in a pore size distribution with lower dispersion and lower mean  
467 values. The results in terms of pore size distribution are in line with similar analyses in the literature,

468 which show that the use of CO<sub>2</sub> in foamed concrete leads to a significant increase in the number of  
469 pores, characterised by better sphericity, more uniform distribution and finer size compared to the  
470 reference plain aerated concrete (Zhang et al. 2024). This distinct microstructure may be explained  
471 by considering the specific physical and chemical characteristics of CO<sub>2</sub>. Carbon dioxide has a higher  
472 molecular mass than air and exhibits significantly greater solubility in water. CO<sub>2</sub> reacts with water  
473 to form carbonic acid, H<sub>2</sub>CO<sub>3</sub>, an unstable compound that, upon reaching a certain concentration,  
474 spontaneously decomposes back into CO<sub>2</sub> and H<sub>2</sub>O. A portion of the released CO<sub>2</sub> diffuses outward,  
475 leading to the formation of new microbubbles (Zhang et al. 2024). This continuous process allows  
476 only fine bubbles to remain stable during the fresh stage, ultimately giving rise to the observed  
477 microstructural features: the use of CO<sub>2</sub> foam resulted in a pore size distribution characterized by  
478 lower dispersion and reduced mean pore size.

479 To further investigate the microstructure of the foamed samples, SEM images were manipulated to  
480 quantify the area occupied by the pores. Contrast and brightness of the images with 50X  
481 magnification were adjusted to obtain white and black pixels, respectively, in correspondence of pores  
482 and paste areas (Figure 14). The area examined for both the FC\_AIR and the FC\_CO<sub>2</sub> samples was  
483 of the same size and therefore had the same number of pixels. The image manipulation software  
484 provided quantification of both white and black pixels. The results of the analysis are shown in Table  
485 3. The ratio of void and paste cross sections to the total area,  $A_{\text{pores}}/A_{\text{tot}}$  and  $A_{\text{paste}}/A_{\text{tot}}$ , is also given.  
486 The analysis shows a larger area occupied by the pores in FC\_CO<sub>2</sub>. In fact, a more uniform  
487 arrangement of the pores within the material corresponds to a more homogeneous size distribution.  
488 As a result, the paste cross section is reduced. The ratio of pore area to the total area increases by  
489 approximately 8% in the FC\_CO<sub>2</sub> sample compared to the FC\_Air sample. This microstructure  
490 contributed to the lower density of the FC\_CO<sub>2</sub> samples compared to the reference (Figure 9).

491 It is worth noting that the microstructure described in terms of pore distribution within the material  
492 may also be related to its flexural behaviour. In fact, the higher presence of pores in the FC\_CO<sub>2</sub>

493 samples may have favoured the initiation of cracks under flexural conditions, resulting in lower  
494 flexural strength.

495 Figure 12 c and d, at intermediate magnification, highlight the struts between the pores for FC\_Air  
496 and FC\_CO<sub>2</sub> samples. A comparison of the two images reveals that the cement paste of the FC\_Air  
497 sample appears to be more compact, despite the presence of micronic pores measuring between 10  
498 and 40 μm. In contrast, the FC\_CO<sub>2</sub> sample exhibits a diffuse porosity characterised by pores in the  
499 submicron to few micron range. This aspect may also have contributed to the higher density observed  
500 in FC\_Air samples. Figure 12 e and f, produced at the highest magnification, highlight the  
501 microstructure of the materials. The presence of portlandite grains, as indicated by XRD and DT-  
502 TGA analysis, can be clearly discerned. Furthermore, while a limited number of calcium carbonate  
503 precipitates can be discerned on the surface of portlandite grains in the FC\_Air sample, a continuous  
504 calcium carbonate structure, distributed all around the portlandite crystals, can be observed in the  
505 FC\_CO<sub>2</sub> sample. This observation can be attributed to the higher carbonation degree that occurs in  
506 the samples produced using CO<sub>2</sub> gas. The presence of flocc-like C-S-H connected to the surrounding  
507 matrix is demonstrated in both images. It is worth noting that the images do not show any evidence  
508 of a more compact pore wall structure associated with calcium carbonate formation. In fact, the  
509 FC\_CO<sub>2</sub> samples still showed a significant portlandite content, highlighting the possibility of further  
510 carbonation of the material. In this respect, the investigation of different carbonation configurations  
511 and their influence on the material microstructure represents a valuable perspective of the study.

## 512 **5. Conclusions**

513 The present study explores the feasibility of producing a novel foamed concrete by using CO<sub>2</sub> gas to  
514 produce the foam, with the aim of inducing carbon capture due to the chemical reaction of carbon  
515 dioxide with fresh cementitious paste. The primary conclusions of the study are outlined as follows:

- 516 - The CO<sub>2</sub> foam has a marked effect on the mineral composition of the foamed concrete,  
517 resulting in a reduction in portlandite content and an increase in calcium carbonate. The  
518 estimated CO<sub>2</sub> uptake was found to be equal to 8.7%, which is almost double the value  
519 observed in common air foamed concrete;
- 520 - The mean compressive strength of the FC\_CO<sub>2</sub> samples was 4.3 MPa, which was  
521 comparable to the value of the reference FC\_Air (5.0 MPa). However, when dry density  
522 values were considered, it was evident that the FC\_CO<sub>2</sub> samples exhibited an excellent  
523 behaviour, with a compressive strength-to-dry density ratio that was comparable to that of  
524 the reference FC\_Air samples.
- 525 - The microstructure analysis of the samples revealed that the CO<sub>2</sub> foam induced smaller and  
526 more homogeneous distributed bubbles compared to the conventional air foam. The latter  
527 was characterised by a relatively higher standard deviation and a greater prevalence of larger  
528 pores, which occupied approximately 30% of the representative analysed area.

529 The preliminary results demonstrate that the production of lightweight foamed concrete through the  
530 utilisation of CO<sub>2</sub> gas represents a promising technique that effectively combines mechanical  
531 efficiency with environmental sustainability. It is evident that, in view of the encouraging outcomes,  
532 there are a number of prospective avenues for further optimisation of the material. Given the higher  
533 density of CO<sub>2</sub> in comparison to air, the target fresh density of 750±50 kg/m<sup>3</sup> could be achieved by  
534 using 6.5 times more CO<sub>2</sub>-generated foam than air-generated foam. This indicates significant  
535 potential for enhancing the performance of CO<sub>2</sub> foam, presenting opportunities to build upon the

536 promising results discussed in this paper. A crucial point emerged with regard to the manufacturing  
537 process. The gas released during the foam generation raises concerns regarding the carbon capture  
538 efficiency. It is therefore evident that the applicability of the process is contingent upon the  
539 implementation of efficacious solutions that can reduce the gas release. This may be achieved, for  
540 instance, by enhancing the foam stability and by avoiding the emission of the gas into the atmosphere.  
541 In this regard, the integration of the process with other industrial plants, which may facilitate the  
542 circulation of the flue gas and by-products, may result in the establishment of a virtuous system whose  
543 overall CO<sub>2</sub> impact results effectively reduced. Therefore, further research is required to optimise the  
544 production process and to reduce the use of foam when using carbon dioxide as a gas, for example,  
545 through the use of viscosity enhancing agents or the use of foaming agents of a different nature.  
546 Another fundamental aspect to be deepened regards the role of the carbonation degree on the physical  
547 and mechanical properties of the foamed concrete. With this regard, research carried out by adopting  
548 CO<sub>2</sub> gas with different concentrations may provide useful insight into the material behaviour.  
549 Finally, the possibility of further carbonating the material paves the way for future investigations  
550 aimed at analysing the influence of curing conditions, for instance in CO<sub>2</sub> environment, on the  
551 microstructure and mechanical properties of the foamed concrete. Furthermore, a more detailed  
552 investigation is necessary to analyse the influence also in terms of thermal and acoustic insulation  
553 behaviour. Additionally, a forthcoming study will delve into properties related to durability, a crucial  
554 aspect in the field of building materials, emphasizing the comparison between the proposed  
555 innovative foamed concrete and conventional foamed concrete.

556

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562

563 **Data Availability Statement**

564 All data, models, or code that support the findings of this study are available from the corresponding  
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713 *Table 1: Chemical composition of cement powder (% wt).*

	Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	Cr <sub>2</sub> O <sub>3</sub>	MnO	Fe <sub>2</sub> O <sub>3</sub>	SrO	LOI
Cem	0.56	1.59	4.51	18.20	0.07	4.44	1.57	63.30	0.19	0.04	0.04	1.73	0.04	3.66

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739 *Table 2: Calcite and portlandite content in hardened samples*

	%CO <sub>2</sub>	%CaCO <sub>3</sub>	%H <sub>2</sub> O <sub>port.</sub>	%Ca(OH) <sub>2</sub>
	[%]	[%]	[%]	[%]
<b>FC_Air</b>	7.0	15.9	3.7	15.1
<b>FC_CO<sub>2</sub></b>	10.9	24.8	2.8	11.5

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764 *Table 3: Quantification of black and white pixels of manipulated SEM images.*

	$\text{pixel}_{\text{tot}}$	$\text{pixel}_{\text{pores,white}}$	$\text{pixel}_{\text{paste,black}}$	$A_{\text{pores}}/A_{\text{tot}}$	$A_{\text{paste}}/A_{\text{tot}}$
				[%]	[%]
<b>FC_Air</b>	766000	476315	289685	62.2	37.8
<b>FC_CO<sub>2</sub></b>	766000	514594	251406	67.2	32.8

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