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CO_2 Foamed Concrete: an innovative approach to combining mechanical strength and carbon storage

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ABSTRACT

Carbon capture and utilization technologies, along with the use of lightweight concrete, offer promising solutions towards the decarbonization of cement industry. Combining both practices, this paper proposes an innovative type of foamed concrete produced with CO_2 gas foam instead of common air foam. The objective is to demonstrate the viability of inducing the formation of porosity in the material, which is necessary for its optimal structure, while simultaneously enabling the permanent storage of carbon dioxide through the chemical reaction between the CO_2 gas and the fresh cementitious paste. Samples are produced using the preformed foaming method, injecting CO_2 gas instead of air, and mixing foam and cement paste prior to casting in prismatic moulds. Common foamed concrete samples produced with air foam are considered as reference.

Compressive tests are carried out to investigate the mechanical properties. X-Ray Diffractometry and Scanning Electron Microscopy are conducted to investigate influence of CO_2 gas on the microstructure of the foamed concrete. Thermogravimetry is adopted to quantify the stored CO_2.

The results obtained from this study are extremely encouraging. The use of CO_2 foam was found to nearly double the CO_2 uptake, without producing any significant change in the compressive strength-to-density ratio. Furthermore, the utilisation of CO_2 foam demonstrated to exert a favourable influence on the microstructure of the material, resulting in the formation of smaller pores that exhibit a more uniform size distribution.

KEYWORDS: CO_2 Foamed Concrete; Compressive strength; Pore size distribution; CO_2 mineralisation.

1.INTRODUCTION

Cement is the principal material adopted in the construction field, due to the global increase in demand for concrete (Mohamad et al. 2022). Consequently, the cement industry is responsible for approximately 7% of global CO_2 emissions (Korczak et al. 2022). In order to comply with the EU climate policy standards, the sector is required to reduce CO_2 emissions by 30% by the year 2030 and to achieve carbon neutrality by the year 2050 (Cembureau 2021). In this regard, strategies for Carbon Capture Storage and Utilization (CCS/CCU) account for 36% of the overall reduction. CCS actions consist of the reduction of greenhouse gas emissions through CO_2 mineralisation processes. CCU is used to denote processes in which carbon dioxide is stored and the products are valuable for a practical use.

Several CO_2 utilisation technologies for construction materials have emerged. These processes involve the reuse of captured CO_2 in the curing, production and modification of building materials (Li et al. 2022). The majority of employed mineralisation techniques include: the use of a carbonation chamber for the curing of pre-cast elements (Ferrara et al. 2021; Librandi et al. 2019); accelerated aqueous mineralisation processes (Bonfante[1] et al. 2024; Bonfante[2] et al. 2024) to produce carbonated powders that are adopted as fillers or cement replacements (Bonfante et al. 2025; Fang et al. 2022).

Recently, CO_2 injection during the mixing process to produce fresh concrete, has emerged as alternative CCU strategy. This novel approach represents a significant departure from the well-established mineralisation processes mentioned above (Ferrara et al. 2024). The formation of calcium carbonate during the fresh mixture resulted in an acceleration of the hydration reactions (Davolio et al. 2025). A limited number of pioneering studies have employed a comparable methodology in order to investigate the potential of utilising CO_2 gas in fresh pastes to develop foamed concrete. In this process, gaseous CO_2 is employed for the generation of bubbles and for reacting with the paste, resulting in the formation of stable calcium carbonates (Ta[1] et al. 2023; Ta[2] et al. 2023).

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

The increasing interest in foamed concrete can be attributed to the straightforward manipulation of porosity, which confers a high degree of versatility in its applications. Indeed, low and medium-low density specimens have been shown to be particularly suited to applications requiring thermal insulation and fire resistance properties. As the density of the material increases, the potential for exploitation as structural materials also grows. This is particularly advantageous in areas of high seismicity (Falliano et al. 2023). Furthermore, foamed concrete is typically distinguished by a high thermal inertia with a time lag comparable to that of conventional materials, yet with a higher decrement, rendering it suitable also for applications in warm environments (Ropelwski et al. 1999).

In recent years, there has been a growing interest in the feasibility of utilising CO_2 as a foaming agent for the production of lightweight foamed concrete attracted interest. In their study, Li et al. 2020, examined the thermal insulation performance of foamed concrete produced with varying foaming gases. The findings of the study indicate that the lower thermal conductivity of CO_2 gas compared to air resulted in the CO_2 foamed concrete exhibiting enhanced thermal insulation compared to traditional foamed concrete. Whilst the objective of the study was not to induce CO_2 mineralisation for carbon storage, it demonstrated the feasibility of producing CO_2 foam.

A number of studies have proposed the preparation of CO_2 foamed concrete by means of a chemical foaming method. This process entails the utilisation of compounds that undergo a reaction with the cementitious paste to produce CO_2 gas, acting as foaming agents (Yao et al. 2022). Examples of this

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

To date, only a limited number of studies have explored the potential for creating foamed concrete using carbon dioxide gas (physical foaming method), with a particular focus on the effects of both mechanical behaviour and carbon storage (Ta[1] et al. 2023; Ta[2] et al. 2023; Liu et al. 2023). In their study, Liu et al. 2023 produced CO_2 modified foam concrete, which exhibiting a notable enhancement in strength compared to the reference sample. Furthermore, the presence of CO_2 gas within the pores was observed to facilitate a greater formation of calcium carbonate. Nevertheless, the assessment of carbon storage was confined to qualitative examinations, lacking the provision of data pertaining to CO_2 uptake. The results obtained were encouraging, with lightened samples having a dry density of 597 kg/m^3, CO_2 uptake of 8.57% and compressive strength of 1.75 MPa (Ta[1] et al. 2023). However, a carbonation pre-treatment was required prior to the addition of CO_2 foam to increase the viscosity of the paste. This process involved the mixing the cement slurry with CO_2 gas for a duration of up to 120 min, with consequent acceleration in the hydration process (Zajac et al. 2020) that may have compromised the development of strength during the curing period. In the study conduced by Zhang et al. 2024, foamed concrete was produced by partially substituting the conventional foam stabiliser (polyacrylic amide) with CO_2. This substitution resulted in enhanced strength when compared to the control samples.

A critical issue associated with the use of carbon dioxide to generate the foam is the release of CO_2 gas into the atmosphere during the process. In this regard, Ta[2] et al. 2023 emphasised the importance of enhancing the stability of the CO_2 foam in order to reduce the gas release. Furthermore, the utilisation of solid waste materials, such as coal gangue and fly ash, is proposed to enhance carbon sequestration capacity, thereby reducing the environmental impact of the process.

The poor stability of CO_2 foam, and its reactivity with portlandite cement, pose significant challenges to the production of CO_2 foamed concrete. The establishment of a clear procedure for the creation of an efficient material is not yet established. The controversial outcomes pertaining to strength, in conjunction with the absence of a precise quantification of the carbonation potential, underscore the necessity for additional investigation to enhance the performance of this innovative material and to delineate its mechanical and environmental capabilities.

In view of the aforementioned considerations, the aim of the present study is to provide a comprehensive evaluation of the potential for CO_2 foamed concrete. In order to achieve this objective, the investigation encompasses the mechanical properties and microstructure of the material, in addition to the evaluation of the stored carbon to define the impact of the process as a CCU technique. The generation of the foam is achieved through the mixing of CO_2 gas with a surfactant, which is then added to the cementitious paste in order to create the foamed concrete. Any pre-treatment is employed to produce cement paste. Results are compared with similar studies from the literature. Notably, no additional substances were introduced to improve the stability of the CO_2 foam, ensuring that the comparison focuses solely on the impact of the foaming gas.

2.RAW MATERIALS

The binder used in the study is Portland cement, and specifically CEM 52.5 R type. The particle size distribution, determined by laser granulometry (Malvern Mastersizer 3000E laser diffraction particle size analyser), exhibited characteristic values d10, d50 and d90 of 1.87 μm, 11.3 μm and 30.6 μm respectively (see Figure 1).

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

The mineralogical composition was investigated by X-ray diffraction analysis (XRD, Empyrean Malvern Panalytical) in the Bragg-Brentano configuration, with Cu-Kα wavelength radiation (0.15406 nm) operating at 40 kV and 40 mA. The 2θ range was from 5° to 70° with an angular step of 0.006°, and this was held for 23 s. The diffractometer was equipped with Soller (0.04 rad), anti-scatter (P7.5), beam mask (10 mm), divergence (¼°) and axial (½°) slits. The XRD spectra are displayed in Figure 2. As expected for cement, the main compounds identified were calcium silicates (belite and alite), aluminoferrites, aluminates and calcium sulphates. The presence of calcium carbonate was also noted.

Thermogravimetric differential thermal analysis (TG-DTA, LabSys evo machine Setaram) was carried out in an N_2 atmosphere at a heating rate of 10 °C/min up to 1050 °C (Figure 3). The curves illustrate the dehydration of gypsum at approximately 140 °C (peak I), the dehydration of portlandite at around 450 °C (peak II), the decomposition of calcium carbonate at approximately 750 °C (peak III). In accordance with the XRD pattern, the minimal mass loss (0.2%) concomitant with portlandite indicates a negligible presence of this compound. The initial CO_2 content, $\%CO_{2,cem}$, was estimated to be 2.3%.

The theoretical maximum CO_2 uptake, as determined by Steinour equation (Huntzinger et al. 2009), was calculated to be 47.8%, based on the XRF elemental composition. This value, which represents the carbonation potential that would theoretically be reached if all the available calcium reacted with

172 CO_2, was utilised to ascertain the carbonation efficiency, η_{carb}, defined as the ratio between the
173 experimental $CO_{2,uptake}$ 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±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.

3.METHODS

3.1 Definition of operational parameters

The study encompasses two series of samples, comprising air and CO_2 foam, respectively. The operational parameters required to produce both the foamed concrete were accurately defined, based on previous studies conducted by the authors. The key factors that must be considered are the pressure of the gas, the concentration of the foaming agent, the mixing rotational speed, water-to-cement ratio, superplasticiser-to-cement ratio, fresh concrete density. This section is dedicated to the presentation of the rationale behind the selection of the parameters.

The production of foam, employing both air and CO_2, was conducted utilising the same foam generator, Figure 4. The operational mechanics of the apparatus are analogous to those exhaustively delineated in Falliano et al. (2021). It has been demonstrated that the drainage of the foam produced is significantly influenced by pressure and the concentration of the foaming agent. Drainage, which is closely associated with the lifetime of the foam and, consequently, to the quality of the foamed concrete produced, is markedly reduced when the pressure used is above 2.5 bar and when the foaming agent concentration is above 3% (Falliano et al. 2021). The pressure employed for foam generation was approximately 3 bar, while the surfactant concentration was selected to be 5%.

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

In both the cases of air and CO_2 foam production, an identical mix proportion was adopted in the creation of the foamed concrete presented in this study. In previous studies (Falliano et al., 2018) and (Falliano et al., 2022), it was established that fresh foamed concrete demonstrated stability across a wide range of solid-to-liquid ratios (i.e., foam + water-to-cement ratio). In consideration of the

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

In light of the higher foam density produced using CO_2 in comparison to that generated using air, a different foam-to-cement ratio was necessitated in each case to achieve the same target density. Specifically, the quantity of CO_2 foam adopted in order to achieve the target fresh density of 750±50 kg/m^3 was precisely 6.5 times greater than the air foam necessary to obtain the same density. In addition to density, liquid drainage, closely linked to foam lifetime (Falliano et al, 2021), proved to be a key distinction between air-generated foam and CO_2-generated foam. Specifically, while air-generated foam exhibited no detectable drainage even ten minutes after preparation, CO_2-generated foam showed measurable drainage within two minutes of its production. The use of CO_2 instead of air results in greater liquid drainage and, consequently, increased foam instability. Indeed, compared to the use of air, CO_2 promotes surface tension-driven drainage and gas diffusion. CO_2 is significantly more soluble in water than air. Utilizing a gas with very low solubility in water slows down gas transport across the aqueous phase compared to a more soluble gas (Fameau et Salonen, 2014). This further explains the greater instability observed in CO_2-based foams compared to those produced with air. Furthermore, the higher instability of CO_2-based foam is also attributed to a greater degree of bubble coalescence, a phenomenon triggered by the higher density of CO_2 compared to air (Liu et al, 2023).This coalescence refers to the physical process by which liquid droplets, gas bubbles, or solid particles merge to form a single larger entity. For this reason, the foam was immediately mixed with the cementitious paste after its generation to prevent the drainage phenomenon. This is a crucial approach for ensuring the production of high-quality foamed concrete.

3.2 Sample preparation and mechanical testing procedures

Foamed concrete samples produced with air foam are designated as FC_Air, while foamed concrete samples produced with CO_2 foam are designated as FC_CO_2. The foam generator was connected to an air compressor in order to generate air foam. In the case of the FC_CO_2 specimens, the generator

was connected to a CO_2 cylinder with gas purity of 99.9% (grade 2.5). Initially, the cement, water, and superplasticiser were mixed until a homogeneous cementitious system was obtained. Immediately following its generation, the requisite quantity of foam was added to the previously prepared cementitious mixture to attain the target fresh density, i.e. 750±50 kg/m^3. The mixing process was continued for a further two minutes or until a homogeneous system was achieved, in any case. Subsequently, the material was accurately poured in order to guarantee optimal adherence to the formwork. Following a period of 48 hours, the prismatic samples, with dimensions of 20 mm x 20 mm x 80 mm, were demoulded and cured in air at room temperature (20±3°C) and relative humidity of 65±5%. The sample size adopted is commonly used in the literature for the characterization of cementitious pastes (Suarez-Riera et al. 2020). For each series, four samples were cast. Three of them were subjected to mechanical testing, while a fourth sample was used for chemical characterisation. Following the curing process, the specimens were subjected to three-point bending tests and compressive tests. These tests were conducted in accordance with the procedure outlined in EN 196-1. The testing procedure involved the application of half of the specified loading rate, calculated based on the scaled sample size. The flexural strength evaluation was carried out in force-controlled mode through a Zwick-Line Z050 testing machine, with a loading rate of 25 N/s, applying the load perpendicularly to the direction of casting. The span length was equal to 60 mm.

The compressive strength was evaluated in force control mode, utilising the Zwick-Line testing apparatus, in addition to the halves of the prisms derived from the three-point bending test. The specimens were subjected to a loading rate of 1200 N/s, applied perpendicularly to the direction of casting. Following the testing phase, the specimens were placed in an oven and dried at a temperature of 100°C, in accordance with standard practice for assessing the density of hardened mixes of cementitious conglomerates. This process was continued for a minimum of 48 hours or until the specimens reached a constant weight, in order to evaluate their dry density. Figure 5 reports a scheme of the process adopted to produce and characterise the foamed samples.

3.3 Carbon capture assessment

This section delineates the methodologies employed for the estimation of the CO_2 chemically bound within the material as a consequence of the formation of calcium carbonate. The efficiency of the carbonation process was evaluated through the analysis of the hardened foamed paste specimens. In particular, a sample was extracted from the core of prismatic specimens 24 hours after casting, milled, and immediately subjected to the analysis. The sample was not subjected to a drying process in order to prevent the additional carbonation of portlandite and the hydration of calcium silicates present in the cement paste, which could occur as a result of prolonged exposure to the atmosphere. XRD and SEM analysis were adopted to qualitatively investigate the formation of calcium carbonate. TG-DTA analysis was adopted to quantify the calcium carbonate content, which is related to the mass loss in the temperature range of 550°C to 850°C (Ferrara et al. 2023). The CO_2 content is expressed as A percentage of the mass with respect to the amount of binder. The latter is considered to be equal to the TG value at a temperature of 950 °C (Mo et al. 2016), after the release of water and CO_2 content deriving from hydration and carbonation reactions (Equation 1):

$$\%CO_2 = \frac{\%\Delta W_{550°C-850°C}}{\%W_{950°C}} \qquad \textit{Equation 1}$$

where $\%\Delta W_{550°C\text{-}850°C}$ denotes the %TG mass loss in the 550°C-850°C range of temperature, and $\%W_{950°C}$ denotes the %TG at 950°C. $\%CO_{2,uptake}$ is then defined as the difference between the $\%CO_2$ and the initial CO_2 content of the raw cement powder, designated as $\%CO_{2,cem}$.

The portlandite content in the hardened samples, $\%Ca(OH)_2$, is determined from TG-DTA curves that exhibit a mass loss due to the release of water during the dihydroxylation calcium hydroxide. In order to eliminate the contribution of C-S-H decomposition from the estimate, the tangent method was adopted (Lothenbach et al. 2016). In addition to the CO_2 content, the portlandite content is also expressed as percentage of the mass at 950 °C.

The TG curve at temperatures below 150 °C was characterised by a significant mass loss, which was 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).

4.RESULTS AND DISCUSSION

4.1 Mineral composition and carbon capture

Figure 6 reports the XRD pattern of FC_Air and FC_CO_2 samples, indicating the main crystalline compounds present in the material. XRD pattern of raw cement is also reported for comparison. As expected, the FC_Air sample displayed the presence of portlandite, a product of calcium silicates hydration, and ettringite, a product of the reaction between gypsum and calcium aluminates. The presence of calcium silicates hydrates, C-S-H, presumed to result from the hydration of belite and alite, was not discernible due to their low crystallinity. Furthermore, peaks attributable to the primary compounds of raw cement, in addition to a number of calcite peaks, were also discerned.

The XRD pattern of the FC_CO_2 sample exhibited analogous compounds to those identified in the FC_Air sample. However, an alteration in the relative intensity of the peaks was observed with an increase in the signal related to calcite in comparison to portlandite. This may indicate, at least qualitatively, that a portion of the portlandite derived from the hydration process reacted with the CO_2 present in the foam, forming calcium carbonates. Furthermore, the XRD pattern exhibited peaks associated with calcium hemicarboaluminate, displaying heightened intensity in comparison to the FC_Air sample. This compound is typically present in the early stages of hydration in Portland cement with calcite content, resulting from the reaction of calcium aluminates. As demonstrated by Ipavec et al. (2011), the substance undergoes a transformation into a more stable monocarboaluminate during periods of prolonged hydration. Consequently, the higher presence in FC_CO_2 samples also corroborates the occurrence of a carbonation process. The presence of mono- and hemicarboaluminates exerts a stabilising effect on ettringite (Zajac et al. 2014). The consequent reduction in the availability of hydrated calcium aluminates (C-A-H) limits its capacity to react with gypsum from sulphatic attack, thus preventing the formation of secondary ettringite.

Figure 7 and Figure 8, respectively, illustrate the TG-DTA curves of the FC_Air and FC_CO_2 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_2.
323 Table 2 presents the data regarding the CO_2 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_2 foam in the FC_CO_2 sample resulted in a
329 reduction in portlandite content, $\%Ca(OH)_2$, 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 $\%CO_2$, of 2.1% should be indicative of this variation in portlandite content. However, the results in
333 Table 1 indicate a higher $\%CO_2$ increase, corresponding to 3.9%. The observed phenomenon can be
334 attributed to the varying reactivity of calcium silicates in the presence of CO_2. 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_2 present in the
339 pores of the FC_CO_2 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 $CO_{2,uptake}$ 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 $CO_{2,uptake}$ of the
345 FC_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 CO_2 foamed concrete, which havs documented
347 CO_2 uptake values ranging from 6.10% to 8.57% (Ta[1] et al. 2023; Ta[2] et al. 2023) and from 4.20% to
348 7.48% (Zhang et al. 2024).
349 The carbonation efficiency, η_{carb}, was found to be equal to 9.9% and 18.1% for the FC_Air and
350 FC_CO_2 samples, respectively. It was observed that the utilisation of 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 CO_2 uptake parameter is used to describe the capacity of the material to
354 permanently store 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 CO_2 balance of the process. A primary and crucial aspect is the release of 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[1] et al. 2023), an eco-efficient process should be integrated with
362 other industrial plants that provide flue gas for use as a 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.

4.2 Mechanical strength

Figure 9 presents a comparison of the dry densities of the two distinct types of foamed concrete under investigation. Indeed, in order to make an effective comparison of the mechanical properties of foamed concrete, it is essential to evaluate its dry density. This methodological approach facilitates more precise comparisons with other studies in the scientific literature, where foamed concrete is characterised by analogous dry densities, thus providing a consistent basis for analysing performance across disparate investigations. The findings of the experiments conducted in order to ascertain the compressive and flexural strengths are presented in Figure 10. Moreover, all the figures present the mean values with standard deviations, alongside the experimental data, in order to provide a quantitative assessment of the dispersion of the findings. While the fresh density is approximately equivalent, the latter figure demonstrates that the discrepancy in terms of dry density is considerably more pronounced. Indeed, samples prepared with CO_2 foam exhibit a reduction in dry density of approximately 15%, which is slightly less than that observed in samples with traditional foam. This is an anticipated outcome, as evidenced by the higher quantity of CO_2 foam, as illustrated in Section 3.1, necessary to attain the target fresh density of 750±50 kg/m^3. It was expected that more CO_2 foam would be needed to achieve the desired target fresh density, since CO_2 foam produced was about 6.5 times denser than air foam. This increased the liquid-to-solid ratio, as reflected in the lower dry density of CO_2-based foamed concrete. However, no coalescence or instability was observed, which can be attributed to the immediate mixing of the foam with the cementitious paste. This stability is attributed to the very low water-to-cement ratio and the carefully designed mix proportions, based on prior expertise (Falliano et al., 2018; Falliano et al., 2022), which ensured stability across a wide liquid-to-solid ratio range, including those used in this study. Despite the non-negligible reduction in dry density, which is contrary to the general expectation (Nambiar et Ramamurthy 2007), the CO_2 foam specimens exhibited a microstructure with voids characterised by smaller diameters and a more homogeneous distribution. This aspect will be outlined in greater detail in Section 4.3. This significant finding enables the ratio of compressive strength, MPa, to dry density, kg/m^3, (σ_c/ρ) 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 kg/m^3, exhibit a ratio of compressive strength to dry density (σ_c/ρ)
394 ranging from 0.0017 to 0.0029 (Falliano et al. 2022). In this study, both for the air and the CO_2 foamed
395 concretes presented in this study, σ_c/ρ 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 kg/m^3 are characterised by a ratio of compressive strength, MPa, to dry density, kg/m^3, approximately
403 equal to 0.006; moving towards 500 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 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-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 CO_2 foamed concrete presented in this
415 study. These excellent results are further substantiated when a comparison is made with the CO_2
416 foamed concrete presented in the study from Ta[1] 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[1] et al. 2023), foamed concretes with densities of 597 kg/m^3 are characterised by
419 a ratio of compressive strength, MPa, to dry density, kg/m^3, equal to 0.0029, while in the case of
420 densities of 621 kg/m^3 and 669 kg/m^3, this ratio increases to 0.003 and 0.0036, respectively.
421 Moreover, in Liu et al. (2023), for samples made with 100% CO_2 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_2 foam, which
424 facilitated the generation of a stable CO_2 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_2, 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_2 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_2 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 (σ_f/ρ). The latter ratio was found to be equal to 0.0027 and
435 0.0018 for air and CO_2 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_2 samples at varying
441 magnifications.

Representative sections of both the FC_Air and FC_CO_2 samples are illustrated in Figure 12a and b, at the lowest magnification, and show the pores generated by the foam during the mixing process that remained in the hardened state. The distribution of pores sizes generated by the foam was subjected to statistical analysis for both samples. A representative continuous region, comprising a population of approximately 2000 pores, was selected for analysis in both the FC_Air and FC_CO_2 samples. The diameter of each pore was then measured. The pores were observed to have a relatively circular shape. In the case of irregular pores, the shortest diameter was deemed to be the most appropriate for consideration. The mean values of pore size were found to be 59.3 μm (St.Dev. 53.7μm) and 52.0μm (St.Dev. 37.7μm), respectively, for the FC_Air and FC_CO_2 samples. To gain further insight into the pore size distribution and to better comprehend the observed variability, the results from the two samples were compared in terms of the area occupied by the pores within the image region of analysis. In order to achieve this objective, the assumption was made that the SEM images were representative of a plane section, and the area of each pore was calculated from the diameter data, assuming a circular shape. The results are presented in Figure 13, where the pore size is reported on the x-axis and the cumulative distribution in terms of area is expressed as a percentage of the total area occupied by the considered pore population on the y-axis.

The curves demonstrate an upper section characterised by irregular, minor steps, indicating a discontinuous distribution due to the presence of pores with markedly larger dimensions. This is followed by a continuous distribution indicating less pronounced difference in size between consecutive data points. The chart demonstrates that the FC_CO_2 sample exhibited a distribution of smaller pores in comparison to the FC_Air sample across the entire population. Moreover, the discontinuous part in the FC_Air curve, which is more pronounced than in the FC_CO_2 curve, indicates that approximatively 30% of the total area is occupied by pores with a size much larger than the average. The statistical analysis confirms the trend that can be seen visually in Figure 12 a and b. In fact, the use of CO_2 foam resulted in a pore size distribution with lower dispersion and lower mean 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_2 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_2. Carbon dioxide has a higher
472 molecular mass than air and exhibits significantly greater solubility in water. CO_2 reacts with water
473 to form carbonic acid, H_2CO_3, an unstable compound that, upon reaching a certain concentration,
474 spontaneously decomposes back into CO_2 and H_2O. A portion of the released CO_2 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_2 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_2 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_{pores}/A_{tot} and A_{paste}/A_{tot}, is also given.
486 The analysis shows a larger area occupied by the pores in FC_CO_2. 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_2 sample compared to the FC_Air sample. This microstructure
490 contributed to the lower density of the FC_CO_2 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_2

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_2 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_2 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_2 sample. This observation can be attributed to the higher carbonation degree that occurs in
506 the samples produced using CO_2 gas. The presence of floc-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_CO2 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.

5.Conclusions

The present study explores the feasibility of producing a novel foamed concrete by using CO_2 gas to produce the foam, with the aim of inducing carbon capture due to the chemical reaction of carbon dioxide with fresh cementitious paste. The primary conclusions of the study are outlined as follows:

- The CO_2 foam has a marked effect on the mineral composition of the foamed concrete, resulting in a reduction in portlandite content and an increase in calcium carbonate. The estimated CO_2 uptake was found to be equal to 8.7%, which is almost double the value observed in common air foamed concrete;

- The mean compressive strength of the FC_CO_2 samples was 4.3 MPa, which was comparable to the value of the reference FC_Air (5.0 MPa). However, when dry density values were considered, it was evident that the FC_CO_2 samples exhibited an excellent behaviour, with a compressive strength-to-dry density ratio that was comparable to that of the reference FC_Air samples.

- The microstructure analysis of the samples revealed that the CO_2 foam induced smaller and more homogeneous distributed bubbles compared to the conventional air foam. The latter was characterised by a relatively higher standard deviation and a greater prevalence of larger pores, which occupied approximately 30% of the representative analysed area.

The preliminary results demonstrate that the production of lightweight foamed concrete through the utilisation of CO_2 gas represents a promising technique that effectively combines mechanical efficiency with environmental sustainability. It is evident that, in view of the encouraging outcomes, there are a number of prospective avenues for further optimisation of the material. Given the higher density of CO2 in comparison to air, the target fresh density of 750±50 kg/m3 could be achieved by using 6.5 times more CO_2-generated foam than air-generated foam. This indicates significant potential for enhancing the performance of CO_2 foam, presenting opportunities to build upon the

promising results discussed in this paper. A crucial point emerged with regard to the manufacturing process. The gas released during the foam generation raises concerns regarding the carbon capture efficiency. It is therefore evident that the applicability of the process is contingent upon the implementation of efficacious solutions that can reduce the gas release. This may be achieved, for instance, by enhancing the foam stability and by avoiding the emission of the gas into the atmosphere. In this regard, the integration of the process with other industrial plants, which may facilitate the circulation of the flue gas and by-products, may result in the establishment of a virtuous system whose overall CO_2 impact results effectively reduced. Therefore, further research is required to optimise the production process and to reduce the use of foam when using carbon dioxide as a gas, for example, through the use of viscosity enhancing agents or the use of foaming agents of a different nature.

Another fundamental aspect to be deepen regards the role of the carbonation degree on the physical and mechanical properties of the foamed concrete. With this regard, research carried out by adopting CO_2 gas with different concentrations may provide useful insight into the material behaviour.

Finally, the possibility of further carbonating the material paves the way for future investigations aimed at analysing the influence of curing conditions, for instance in CO_2 environment, on the microstructure and mechanical properties of the foamed concrete. Furthermore, a more detailed investigation is necessary to analyse the influence also in terms of thermal and acoustic insulation behaviour. Additionally, a forthcoming study will delve into properties related to durability, a crucial aspect in the field of building materials, emphasizing the comparison between the proposed innovative foamed concrete and conventional foamed concrete.

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Data Availability Statement

All data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

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Table 1: Chemical composition of cement powder (% wt).

	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	K_2O	CaO	TiO_2	Cr_2O_3	MnO	Fe_2O_3	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

Table 2: Calcite and portlandite content in hardened samples

	$\%CO_2$ [%]	$\%CaCO_3$ [%]	$\%H_2O_{port.}$ [%]	$\%Ca(OH)_2$ [%]
FC_Air	7.0	15.9	3.7	15.1
FC_CO_2	10.9	24.8	2.8	11.5

763

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

	$pixel_{tot}$	$pixel_{pores,white}$	$pixel_{paste,black}$	A_{pores}/A_{tot} [%]	A_{paste}/A_{tot} [%]
FC_Air	766000	476315	289685	62.2	37.8
FC_CO_2	766000	514594	251406	67.2	32.8

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