

# Concrete carbon mixing – A systematic review on the processes and their effects on the material performance

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## ABSTRACT

The need for concrete industry to meet the climate neutrality target raised the attention towards carbon capture and utilization (CCU) technologies. Among the various solutions, cementitious materials can benefit from enforced carbonation, a thermodynamically stable reaction that permanently sequesters carbon dioxide into cement compounds. A possible enforced carbonation process is concrete carbon mixing, which involves the addition of pumping systems into concrete production lines to inject carbon dioxide into the material while in a fresh state. Various studies attempted to improve the efficiency of the process and to increase the quantity of fixed carbon dioxide. The current literature was systematically analysed to provide an overview of process parameters, possible injection systems, and properties of carbonated cementitious products. The studies were classified according to the injection stage: carbonation of the mixing water, carbonation of the cement slurry, or injection during concrete mixing with all the components. Concrete carbon mixing has proven to be promising for carbon dioxide sequestration through enforced carbonation, as the injection process enhanced the properties of the final product in most instances. In addition, other relevant aspects of carbon dioxide sequestration processes were discussed. Firstly, the methods and formulations to determine the CO<sub>2</sub> uptake were presented together with cross-comparison studies. Moreover, the methodological aspects of life cycle assessment (LCA) applied to concrete carbon mixing processes were discussed, showing the lack of systematic studies. In conclusion, simplified evaluations demonstrated the economic viability of carbon dioxide injection in fresh concrete, supporting future industrial deployment and discussing the challenges for the upscaling.

## 1. Introduction

The progressive increase of the average temperature of planet Earth raised concerns towards the significant anthropic greenhouse gas emissions. The urgent need for solutions to reduce the Global Warming Potential (GWP) – the main parameter to assess the effects of carbon dioxide emissions on the global temperature – encouraged political, social, and industrial stakeholders to identify and address the required changes in the most emitting fields. For instance, the Global Cement and Concrete Association (GCCA) developed a roadmap to cut the carbon dioxide emissions of the concrete industry (Lehne and Preston, 2018; Le Quére et al., 2016) down to net zero within 2050, meeting the objectives set by the European Commission for a climate-neutral European Union (i.e., net-zero greenhouse gas emissions) through the European Climate Law, which aims to pursue the global commitment reached with the Paris Agreement in 2015 (Cement et al., 2050). Similar targets were set by the European Cement Association (CEMBUREAU) through the “5C” strategy (Cembureau, 2020; Cembureau, 2024). According to the 2006 Intergovernmental Panel on Climate Change (IPCC) Guidelines (Hanle et al., 2006), the emission factor for Portland cement clinker is 0.52 ton<sub>CO2</sub>/ton<sub>clinker</sub>, which only accounts for the CO<sub>2</sub> released by the

calcination reaction. Assuming that the increase both in the world population and in the demand of housing and infrastructures in growing and emerging countries will still continue in the next decades and, considering the lack of true alternatives to cement-based materials to economically satisfy such large demand, the cement industry has put increasing efforts in the recent years to explore alternative solutions to conduct the calcination process with more sustainable thermal energy sources. Nonetheless, as long as traditional clinker is produced, the calcination emissions cannot be avoided; thus, they must be compensated to fulfil the net-zero goal for the cement and concrete industrial production. In the past years multiple technologies were developed both at laboratory and industry level to implement carbon capture, utilization, and storage (CCUS) solutions in the construction industry.

Once carbon dioxide is captured, it is necessary to identify proper feedstocks to permanently store it, preventing further release in the atmosphere. Hence, the feedstocks must be thermodynamically stable and feature a long service life. The major driver for the CCUS technologies is the mineralization reaction, that involves carbon dioxide gas and alkaline elements such as calcium (Ca) and magnesium (Mg). When carbon dioxide is dissolved in water, it can react with the alkaline phases to form stable magnesite and calcite rocks. The abundance of calcium and

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magnesium bearing materials provides a significant opportunity for the long-term storage of the captured CO<sub>2</sub> (Gadikota, 2021). Together with the natural mineral deposits, multiple anthropic sources can be used, such as industrial residues (e.g., slags) and tailings. Thonemann et al. (Thonemann et al., 2022) provided a comprehensive framework for mineralization technologies, with specific regards towards ex-situ carbon capture and utilization, as summarized in Fig. 1. In-situ carbonation involves the injection of carbon dioxide in underground cavities to react with the alkalis present on the site for permanent fixation (Snæbjörnsdóttir et al., 2020). Conversely, ex-situ carbonation is defined as the carbonation of a pretreated feedstock above ground (e.g., in a plant) to produce a value-added material for specific use. Carbonation is a two-step process that requires an initial dissolution of both the carbon dioxide and the metal ions, that subsequently react to form carbonates (e.g., MgCO<sub>3</sub>, CaCO<sub>3</sub>). Owing to their high alkalinity, cementitious materials are valuable carbonation feedstocks, as well as other waste materials commonly used as supplementary cementitious materials (SCMs) for concrete production, including steel slags and fly ashes. Bobicki et al. (Bobicki et al., 2012) described the two main carbonation routes for alkaline minerals, being the direct – or one-step – carbonation and the indirect – or two-step – carbonation. The former involves the carbonatable feedstock and the carbon dioxide that directly reacts to form the carbonated product, either magnesium or calcium carbonate. The latter requires an additional step to extract the alkaline ions (Mg<sup>2+</sup>, Ca<sup>2+</sup>) that then react with CO<sub>2</sub> to carbonate. Further details on the direct carbonation were provided by Zajac et al. (Zajac et al., 2023), defining the dry, semi-dry, and wet processes based on the environmental conditions (i.e., temperature, pressure) and the presence of water during the carbonation reaction.

A relevant parameter to address carbonation reactions is the time-frame over which they occur. Being a thermodynamically favourable reaction, carbonation naturally occurs when alkaline minerals are exposed to carbon dioxide. The environmental conditions such as temperature, CO<sub>2</sub> concentration, and relative humidity govern the kinetics of the process, which is based on the diffusion of the gas through the mineral (Chen et al., 2018). Natural carbonation – also referred to as

weathering carbonation – is characterized by a slow reaction rate, occurring over years or geological eras. This process can be artificially accelerated mimicking the natural reactions in controlled environments, a process called accelerated carbonation. It is however necessary to underline that some differences stand between natural and accelerated carbonation processes, such as the formation of bicarbonates (Auroy et al., 2018). In opposition to natural and accelerated carbonation, enforced carbonation is an engineered process specifically designed to foster the carbonation reaction in material feedstocks that are meant to become valuable industrial products. Enforced carbonation can be used to valorise, for instance, recycled concrete, steel slags, or fly ashes (Zajac et al., 2021; Chen et al., 2021; Ćwik et al., 2018). An example of enforced carbonation for cementitious materials is the process of concrete carbonation curing, which consists of exposing a concrete manufacture to a high CO<sub>2</sub> concentration atmosphere to accelerate the hardening process as well as to enhance the mechanical properties of the final product. Concrete carbonation curing is conducted in sealed chambers with high carbon dioxide concentrations and, eventually, high pressures (up to 4 bar) (Klemm and Berger, 1972; Liu and Meng, 2021). Compared to weathering carbonation, the enforced processes can achieve only limited amounts of carbon dioxide sequestration, but the time required is remarkably reduced. For instance, estimations of the CO<sub>2</sub> uptake due to natural carbonation based on the second Fick's law yielded values ranging between 6 % and 24 % of the total calcination emissions over 40 and 70 years of service life respectively (Pade and Guimaraes, 2007). On the other hand, the CO<sub>2</sub> uptake achieved by Liu et al. (Liu et al., 2022) through concrete carbonation curing was between 1.6 % and 3.7 % by weight of cement, thus comparable to some of the results obtained for natural carbonation, but within 12 h. In addition, early-stage carbonation only involves a limited part of the carbonatable binders and hence further uptake can happen during the service life of carbonated concrete products, at a slightly lower rate (Zhang et al., 2020).

Together with carbonation curing, which is characterized by the exposure of precast concrete manufactures to a CO<sub>2</sub>-rich environment during the hardening process, another technology was recently developed to bind carbon dioxide to fresh concrete, namely carbonation

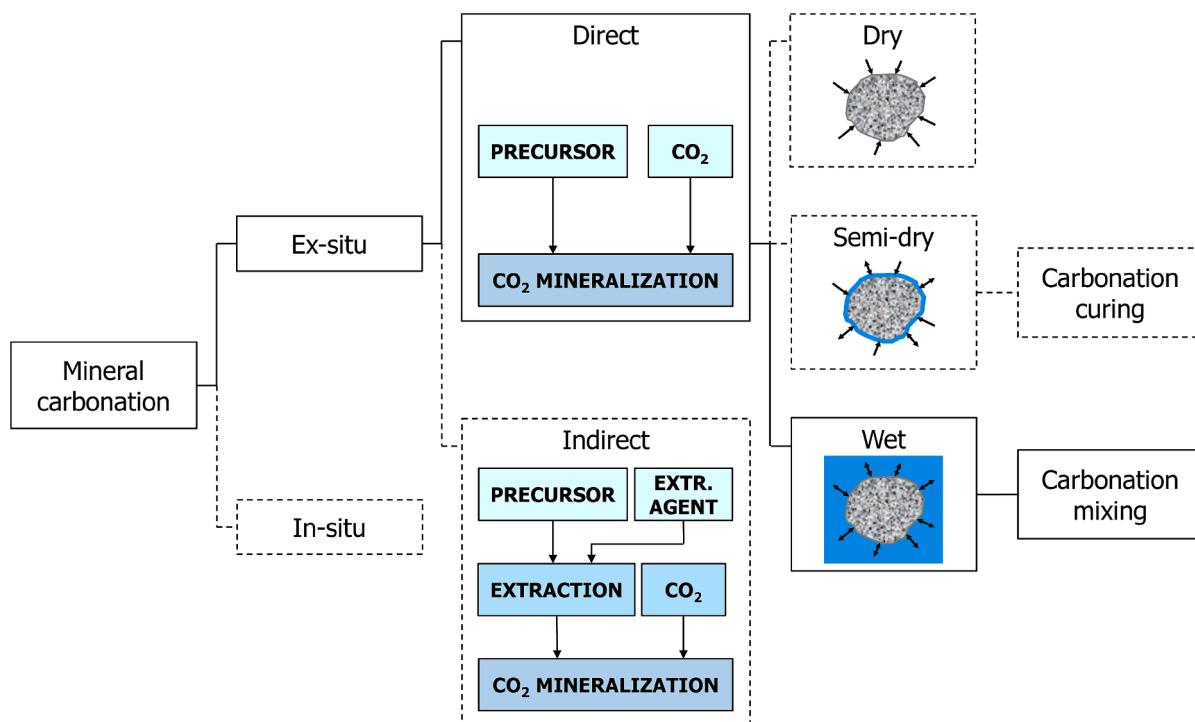
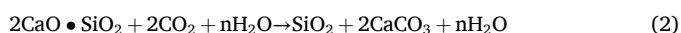
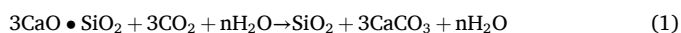


Fig. 1. Framework of the mineral carbonation processes (adapted from (Thonemann et al., 2022; Bobicki et al., 2012; Zajac et al., 2023).

mixing. According to the definition provided by Thonemann et al. (Thonemann et al., 2022), concrete carbonation mixing involves the injection of high-purity carbon dioxide gas during the mixing process of concrete production, when the components are being mixed (He et al., 2017). The concrete carbonation mixing process can both target the cement slurry (i.e., the solution with water and cementitious binder) or the fresh concrete, which includes additives and aggregates. In all cases, the process is conducted in presence of water and not on the dry powder, and the feedstock is in the fluid state. The fresh mixture reacts with the CO<sub>2</sub> to precipitate nano-CaCO<sub>3</sub> particles, with multiple effects on the subsequent hydration and compressive strength development, as it will be further discussed in the upcoming sections.

Concrete carbonation mixing relies on the capacity of the concrete mix constituents to bind carbon dioxide owing to their calcium bearing phases. There are multiple works available in literature that provided a broad overview of the reactions occurring during the carbonation of concrete at fresh state (Zajac et al., 2023; Savija and Luković, 2016; Li et al., 2023). The carbonation processes involve both the hydrated – portlandite (Saetta et al., 1993), C-S-H gel (Peter et al., 2008; Morandeanu and White, 2015; Liu et al., 2022), ettringite (Nishikawa et al., 1992) – and unhydrated phases, thus affecting the final composition of the hardened material and the strength development due to the hydration process (Hu et al., 2022). Together with the carbonates, the carbonation reaction in CO<sub>2</sub>-rich environments yields different products depending on the reaction conditions (Wang et al., 2022), such as the solution pH and the presence of alkalis (De Weerd et al., 2019; Zajac et al., 2021).

The carbonation of unhydrated phases (Ashraf and Olek, 2016) is particularly relevant for concrete carbon mixing since the interaction between the cementitious feedstock and the carbon dioxide gas happens during the initial (i.e., dormant) stage of hydration, when most of the hydrates are not formed yet. Specifically, both tricalcium silicate (C<sub>3</sub>S) and dicalcium silicate (C<sub>2</sub>S) have a fast reaction rate, within the first 10 min of carbonation, while tricalcium aluminate (C<sub>3</sub>A) and tetra-calcium alumina ferrite (C<sub>4</sub>AF) have limited reactivity (Zajac et al., 2020). The reaction of C<sub>3</sub>S and C<sub>2</sub>S with CO<sub>2</sub> in presence of water (Equation (1), Equation (2)) leads to the formation of calcium carbonate together with silica gel as the two main products, as observed by Goodbrake et al. (Goodbrake et al., 1979) and by Groves et al. (Groves et al., 1990).



Finally, although most studies focused on the effects of early enforced carbonation on the phase composition of the hardened cement paste, the hydration products also govern the properties of the interfacial transition zone (ITZ) (Kishore and Tomar, 2023). Currently, the literature covering the investigation of ITZ properties for concrete subject to enforced carbonation is limited to the carbonation pre-treatment of recycled concrete aggregates (RCA) to enhance their interfacial properties (Zhang et al., 2015; Fang et al., 2021; Li et al., 2021). However, the enforced carbonation of recycled concrete aggregates is a separate process and thus it is outside the scope of the present review.

A thorough evaluation of the net environmental benefits associated with carbon curing and carbon mixing was conducted by Ravikumar et al. (Ravikumar et al., 2021). Although the outcomes questioned the effectiveness of such processes as carbon dioxide sinks, the investigated datasets covering concrete carbon mixing were limited to the few works available in literature, with contradictory results. Nonetheless, the compressive strength of the carbonated concrete increased in 20 out of 29 cases, with a maximum observed uptake of approximately 4 kg CO<sub>2</sub> per m<sup>3</sup> of concrete within the investigated datasets. Hence, the carbon mixing pathway can be considered a promising route for carbon capture and uptake in the concrete industry.

From an environmental perspective, the recently published EN

15804 + A2 Standard (Committee, 2019) included the greenhouse gases (GHG) removal from carbonation of cement-based materials in the calculation of the fossil global warming potential (GWP-fossil). In addition, the timeframe for CO<sub>2</sub> sequestration can play a crucial role for the consequent effects of the global warming potential variation on the climate change. Alternative life-cycle assessment (LCA) methods tried to address the role of sequestration time through dynamic-GWP models (Levasseur et al., 2010), emphasizing the relevance of the early-stage sequestration compared to slower processes such as weathering carbonation (Moro et al., 2022). Therefore, enforced carbonation processes gained traction in the concrete industry owing to the significant emissions reduction imposed by the sustainability policies.

Current challenges for concrete carbon mixing technologies include their limited sequestration capacity relative to the emissions from Portland cement clinker production, as well as uncertainties regarding performance outcomes in fresh state, hardened state, and durability properties. Additionally, the early carbonation of unhydrated phases might hinder the microstructural development of concrete, thus leading to poor mechanical performance of the material, associated with C-S-H gel formation. In addition, limited precipitation of portlandite or its excessive transformation into calcium carbonate can result in insufficient alkalinity within the material. This reduction in alkalinity poses a substantial risk to the protection of steel reinforcement by compromising the passivation layer.

The present review aims at providing a systematic assessment of the current developments in concrete carbon mixing. Section 2 briefly describes the quantification and calculation methods for the most significant carbon sequestration metrics. Section 3 highlights the methodology adopted for the review, including the criteria for the selection of suitable studies, the industrial process-oriented classification system, and the source of CO<sub>2</sub> used. The results of the review are presented in Section 4. The latter includes a detailed overview of the three main approaches for industrial implementation of concrete carbon mixing. The most relevant parameters for each system are identified and critically discussed together with the effects of enforced carbonation on the properties of the final products. Section 5 provides the crucial methodological aspects for the life cycle assessment (LCA) of carbonated concrete. Finally, Section 6 delves into the challenges for the scale-up of concrete carbon mixing from laboratory trials to industrial deployment, substantiated by the limited economic data currently available in literature.

## 2. Carbon dioxide uptake assessment

The quantity of carbon dioxide fixed inside the final product through the enforced carbonation techniques is crucial for the evaluation of the effectiveness of such processes towards sustainable concrete production. On the one hand, the CO<sub>2</sub> uptake is a governing parameter for the development of the concrete properties such as the compressive strength and the workability. Therefore, a correlation between the input value (i.e., the CO<sub>2</sub> uptake) and the resulting output can be established only if reliable and repeatable measurements are conducted on the carbonated product to determine the actual uptake. Moreover, the sequestration of carbon dioxide plays a crucial role in the environmental assessment of CCU processes (Hanle et al., 2006). Hence, the effects of inaccurate CO<sub>2</sub> uptake calculations can significantly alter the results of the life cycle assessment, with specific regards to the global warming potential (GWP) impact category (Sacchi and Bauer, 2020). The European guidelines for the Environmental Product Declarations (EPDs) (Committee, 2019) recently adjusted the rules to account for the role of carbonation of cement-based materials in the calculation of the fossil-GWP. Specifically, greenhouse gas removals enabled by concrete carbonation can now be considered. Further details are provided in the product category rules for concrete in the European Standard EN 16757:2022 (Committee, 2022), where the characterization factor of carbon dioxide fossil removed through carbonation is  $-1 \text{ kgCO}_2\text{-eq}$  (Annex C). Additionally, Annex G provides the general background for CO<sub>2</sub> uptake

calculations for each stage of the life cycle assessment. The maximum theoretical uptake depends on the reactive calcium oxide (CaO) available in the binder, as follows:

$$U_{\text{tec}} = w \times C \times (m_{\text{CO}_2}/m_{\text{CaO}}) \quad (3)$$

Where  $w$  is the ratio between the mass of reactive CaO and the mass of binder,  $C$  is the mass of binder, and  $m_{\text{CO}_2}$  and  $m_{\text{CaO}}$  are respectively the molar masses of carbon dioxide (44 g/mol) and calcium oxide (56 g/mol). Based on EN 16757, which reports an average reactive CaO content of 65 % in clinker, and assuming an average clinker content of 95 % in Portland cement, the maximum theoretical CO<sub>2</sub> sequestration is estimated to be 0.49 kg CO<sub>2</sub> per kg of clinker (Committee, 2022). Nonetheless, under atmospheric conditions the carbonation potential is hardly reached. In contrast, most researchers refer to the maximum theoretical uptake including all the mineral phases, as reported in the Steinoor formula (Equation (4)) (Ashraf and Olek, 2016; Steinoor, 1959; Martín et al., 2019; Sanjuán et al., 2020; Huntzinger et al., 2009; Huntzinger et al., 2009).

$$\text{CO}_{2,\text{th}} = 0.785(\text{CaO} - 0.7\text{SO}_3) + 1.091\text{MgO} + 1.42\text{Na}_2\text{O} + 0.935\text{K}_2\text{O} \quad (4)$$

The Steinoor formula relies on the assumption that all the calcium, magnesium, sodium, and potassium oxides will react to form their respective carbonates (i.e., CaCO<sub>3</sub>, MgCO<sub>3</sub>, Na<sub>2</sub>CO<sub>3</sub>, and K<sub>2</sub>CO<sub>3</sub>). Souto-Martinez et al. (Souto-Martinez et al., 2017) developed a simplified mathematical model to predict the carbonation potential with respect to the cement type. However, the model relies on the assumption that the only reactive hydrate phase is CH, neglecting the contributions of C-S-H and aluminates. The EN 16757:2022 Standard (Committee, 2022) also introduced the degree of carbonation, defined as the part of the CaO that has converted to CaCO<sub>3</sub>. For concrete manufactures, the degree of carbonation depends on the carbonation depth achieved at a given time, based on the rate of carbonation  $k$  (Technical Committee, 2019). Nonetheless, the concept of carbonation degree needs to be adjusted for early-stage carbonation processes, since it does not depend on a linear diffusion process, and it shall be determined experimentally instead. Therefore, the key performance index (KPI) generally adopted to assess the performance of enforced CO<sub>2</sub> sequestration processes is the carbon dioxide uptake (CO<sub>2,uptake</sub>). There is no general agreement in literature on the formulation to calculate the CO<sub>2</sub> captured (CO<sub>2,uptake</sub>). The most used formula is reported in Equation (5), and it includes the carbon dioxide absorption due to carbonation (CO<sub>2,bound</sub>) and a reference mass, usually the powder or binder used as capture medium (M<sub>binder</sub>).

$$\text{CO}_{2,\text{uptake}}(\%) = \frac{\text{CO}_{2,\text{bound}}}{M_{\text{binder}}} \times 100 \quad (5)$$

The measurements of the CO<sub>2</sub> bound in a feedstock through the carbonation reaction is not trivial. There are numerous proposed methodologies in the literature, based alternatively on direct or indirect measurements. Some analytical techniques were derived from the assessment of the natural carbonation occurring on concrete manufactures (Qiu, 2020). The quantification of the carbon dioxide fixed through enforced carbonation processes requires refined techniques.

The most diffused technique (Fig. 2) is the thermogravimetry and differential thermal analysis (TG-DTA), which is based on the mass variation under progressive temperature increase (Kikuchi and Kuroda, 2011; Andrade and Sanjuán, 2021; Galan et al., 2012; Scrivener et al., 2016). Although being frequently adopted, the TG-DTA is susceptible to statistical uncertainty due to the limited volume of material analysed (5–50 mg), particularly for samples collected from concrete batches. Therefore, the chemical analysis of the sample can provide further information on the composition and, thus, increase the representativeness of the test (Villain et al., 2007). Furthermore, the presence of aggregates in the ground sample might strongly alter the results due to the possible

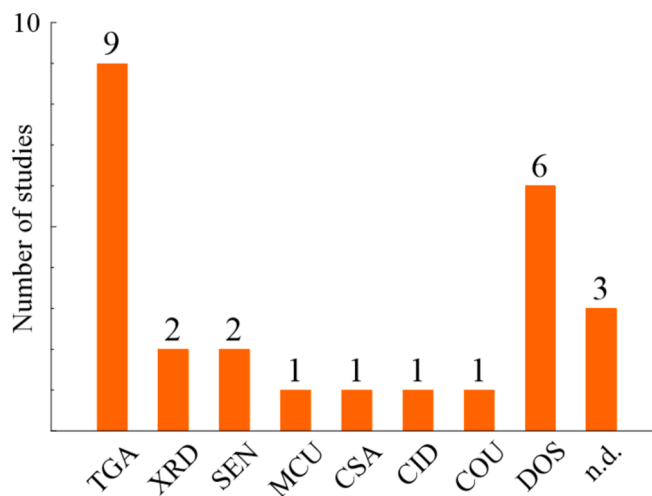


Fig. 2. CO<sub>2</sub> uptake measurements used in the studies included in the review; TGA: Thermogravimetric analysis, XRD: Quantitative X-ray diffraction, SEN: Indirect with CO<sub>2</sub> sensor, MCU: Mass curve, CSA: Total carbon/sulphur analyser, CID: Combustion infrared detection, COU: Coulometer, DOS: Only dosage reported.

presence of calcite. Hence, if TG-DTA is conducted on concrete samples, it is necessary to adjust the measured CO<sub>2</sub> content according to the aggregates' composition, as reported by Ferrara et al. (Ferrara et al., 2023). Other characterization methods include the use of a muffle furnace coupled with weight measurements at given temperatures (Mancarella et al., 2022), mass-based methods that measure the variation of the mass of the sample due to the carbonation process, either as a difference (mass gain) (Zhan et al., 2014; He et al., 2019), or continuously recorded by a scale (mass curve) (Rostami et al., 2012; Shao and Morshed, 2015), acid-base titration (Groves et al., 1990; Belcher et al., 1958; Takahashi et al., 2023; Chang and Y., 2002; Monkman et al., 2018) through reaction with hydrochloric acid (HCl), either at constant atmospheric pressure (Sadallah et al., 2007), or at constant volume (Müller and Gastner, 1971). Moreover, analytical techniques generally adopted to determine the content of each compound in cementitious samples can also be used to determine the carbon dioxide uptake. These techniques include the Quantitative X-Ray Diffraction (Q-XRD) with Rietveld refinement (Fernández-Carrasco et al., 2012; Tu et al., 2016; Wang et al., 2021), the Fourier Transform Infrared Spectroscopy (FTIR) (dos Santos et al., 2021), and the Raman spectroscopy (Kontoyannis and Vagenas, 2000; Mi et al., 2021; Mi et al., 2024). It is however necessary to highlight that in some cases the Q-XRD measurements can exhibit significant differences from other methods such as TGA due to the presence of amorphous phases that cannot be detected (Teune et al., 2023). Finally, total inorganic carbon (TIC) can be indirectly calculated as a difference between the total carbon (TC) and the total organic carbon (TOC) determined by combustion-infrared absorption with a total carbon analyser (He et al., 2017; Higuchi et al., 2014; Monkman, 2018). Alternatively, CO<sub>2</sub> absorption due to carbonation can be calculated indirectly by measuring the variation of carbon dioxide concentration from a given initial concentration value (Boumaaza et al., 2020; van Tonder and Low, 2021; Kwasny et al., 2014), or the gas flow in the carbonation chamber via flowmeter (Liu et al., 2021). The sample volume that can be analysed with each technique is particularly relevant for the representativeness. Fig. 3 summarizes the average sample size for the analytic methods described above. The ratio between the sample size and batch size is particularly important and it is influenced by the type of specimen from which the sample is taken. For instance, concrete presents significantly greater heterogeneity compared to cement paste, affecting both the consistency and reliability of the sample.

Various attempts were made to assess the reliability of each CO<sub>2</sub>



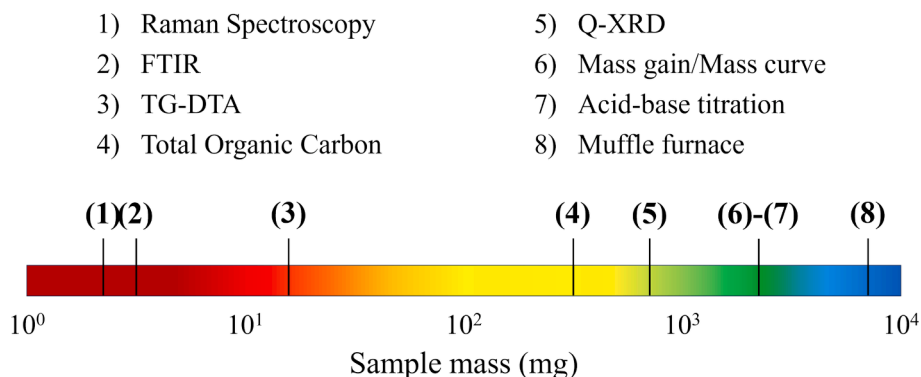


Fig. 3. Range of sample volumes.

quantification method with cross-comparison of different techniques. Rostami et al. (Rostami et al., 2012) measured the CO<sub>2</sub> uptake both with mass-gain and mass-curve methods on an ordinary Portland cement (OPC) paste. A similar comparison was proposed by Shao and Morshed (Shao and Morshed, 2015), with the additional contribution of a thermal furnace treatment. A flask was placed on the scale for the mass-based methods to collect the water evaporated during the treatment. The mass gain method was reported to be an underestimation of the actual uptake due to vapor loss, while mass curve included the water collected through the flask. Ferrara et al. (Ferrara et al., 2023) investigated the carbon dioxide uptake using three different methods, the TG-DTA, the thermal decomposition in the muffle furnace, and the chemical decomposition with hydrochloric acid. The feedstock considered for the carbonation process was an oxygen furnace steel slag. Takahashi et al. (Takahashi et al., 2023) compared the barium carbonate back titration, the thermogravimetric analysis, and the combustion-infrared absorption method with the total carbon analyser. The carbon dioxide uptake was determined for various concrete binders and aggregates. The indirect measurement method developed by Boumaaza et al. (Boumaaza et al., 2020) was validated by comparing the results with TGA measurements on synthesized anhydrous and hydrated cement phases. The FastCarb project (Torrenti et al., 2022) developed a guideline document including the recommendation concerning the CO<sub>2</sub> uptake measurement. The guidelines were drafted according to a cross-laboratory study that assessed the accuracy and repeatability of calcimetric methods, thermogravimetric analysis, and LOI measured with an instrumented furnace. The results indicated the best repeatability for the furnace measurements, with a variation between 1 and 3 kgCO<sub>2</sub>/ton of powder (0.1–0.3 % CO<sub>2</sub> uptake). Comparable results were obtained by the

calcimetric measurements, with an average uncertainty of 0.21 %, while TGA exhibited a slightly higher scattering (4 kgCO<sub>2</sub>/ton or 0.4 %) (Torrenti, 2024). Table 1 summarizes the variation among the different methods as assessed in the comparative studies. The coefficient of variation within the same material stands below 10 % for the studies that used at least three different assessment methods (Ferrara et al., 2023; Shao and Morshed, 2015), while it is slightly higher when only the two mass-based methods are applied (Rostami et al., 2012). The significant scattering observed for blended cements by Takahashi et al. (Takahashi et al., 2023) were associated with the oxidation of sulphides for the ground granulated blast furnace slag (GGBS) and the combustion of unburned carbon for fly ash (FA) that respectively underestimate and overestimate the CO<sub>2</sub> uptake measured by TGA. Moreover, the overestimations of the total carbon by the carbon/sulphur analyser were attributed to the interference of the sulphur on the carbon measurements.

### 3. Methods for the systematic process-oriented review

#### 3.1. Systematic literature review

The present work provides a systematic literature review on concrete enforced carbonation during the mixing stage, following the methodology proposed by A. Fink (Fink, 2014). The purpose of the review is to address the effects of concrete carbonation mixing on the main properties of the final concrete product, including rheological, mechanical, and durability performance. Multiple scientific databases were searched to collect the articles for the systematic review, including Scopus, ScienceDirect, Springer Link, and Wiley. The works included in the

Table 1

Summary of the comparative CO<sub>2</sub> uptake assessments and coefficient of variation (CoV) within each material.

| Study                    | Material     | CO <sub>2</sub> uptake (%) |            |        |               |         |               |              | CoV  |
|--------------------------|--------------|----------------------------|------------|--------|---------------|---------|---------------|--------------|------|
|                          |              | Mass gain                  | Mass curve | TG-DTA | Chem. decomp. | Furnace | HCl back tit. | Total carbon |      |
| (Rostami et al., 2012)   | OPC paste    | 7.6                        | 8.9        |        |               |         |               |              | 0.11 |
|                          |              | 7.6                        | 8.9        |        |               |         |               |              | 0.11 |
|                          |              | 7.5                        | 8.9        |        |               |         |               |              | 0.12 |
| (Shao and Morshed, 2015) | OPC concrete | 13.0                       | 15.2       |        |               | 14.5    |               |              | 0.08 |
|                          |              | 11.2                       | 13.4       |        |               | 12.6    |               |              | 0.09 |
| (Ferrara et al., 2023)   | BOFSS powder |                            |            | 17.5   | 16.5          | 16.0    |               |              | 0.05 |
|                          | BOFSS paste  |                            |            | 13.9   | 13.6          | 13.4    |               |              | 0.02 |
|                          | BOFSS mortar |                            |            | 14.3   | 13.9          | 14.9    |               |              | 0.03 |
| (Takahashi et al., 2023) | OPC powder   |                            |            | 2.09*  |               |         | 2.21*         | 2.14*        | 0.03 |
|                          |              |                            |            | 1.39*  |               |         | 1.47*         | 1.56*        | 0.06 |
|                          |              |                            |            | 2.00*  |               |         | 2.09*         | 2.13*        | 0.03 |
|                          |              |                            |            | 1.40*  |               |         | 1.40*         | 1.47*        | 0.03 |
|                          |              |                            |            | 1.40*  |               |         | 1.52*         | 1.49*        | 0.04 |
|                          |              |                            |            | 2.17*  |               |         | 2.26*         | 2.25*        | 0.02 |
|                          |              |                            |            | 0.27*  |               |         | 0.41*         | 0.43*        | 0.24 |
|                          |              |                            |            | 0.24*  |               |         | 0.18*         | 1.47*        | 1.16 |

\* CO<sub>2</sub> mass (%) in the sample, not CO<sub>2</sub> uptake

literature search were defined addressing the keywords “concrete AND carbonation AND mixing”, specifically targeted to determine applications of the technology both at laboratory and industrial scale. The “concrete AND carbonation AND mixing” keywords provided a total of 12,748 articles. A preliminary screening on the articles limited the research to the peer-reviewed papers written in English. No specific restrictions were adopted concerning the date of publication. Further methodological screening was done following these criteria:

- To be included in the review, the article must contain experimental work conducted by the authors of the article itself and not published elsewhere;
- To be included in the review, the article must provide a description of the carbonation process used to produce the samples.

The refined and final selection of the articles featured a quality assessment of the data provided. Firstly, experimental data must include statistical analyses (e.g., standard deviation, average and extreme values). In addition, the experimental work must focus on the use of carbon dioxide mixed within the production line of concrete, excluding cast concrete (i.e., carbonation curing).

The systematic review includes a total of 22 relevant articles (Fig. 4).

### 3.2. Process-oriented classification

The present review identified three alternatives for the injection of the carbon dioxide. Although all relying on the same basic principles of concrete carbonation previously described, these different approaches are particularly relevant for the eventual upscaling at industrial level. The classification was developed according to the steps of the traditional concrete mixing streamline and considering the retrofitting of the existing equipment by implementing the injection device and/or by adjusting the current instruments.

Firstly, the carbon dioxide can be injected into a sealed water tank (Fig. 5a). The carbonated water is then discharged into the mixer together with the other constituents of the mix. Similarly, the injection can be implemented in a sealed mixer where the cementitious binder is delivered and mixed with water (Fig. 5b). The other constituents of the mix, such as the aggregates or eventual admixtures, can be delivered into the mixer afterwards, thus enabling to control the initial carbonation process before forming the final product. In the second case, an alternative approach would be to inject carbon dioxide into only a portion of the final mix, such as a slurry containing half of the cement specified in the mix design. However, none of the studies reviewed have

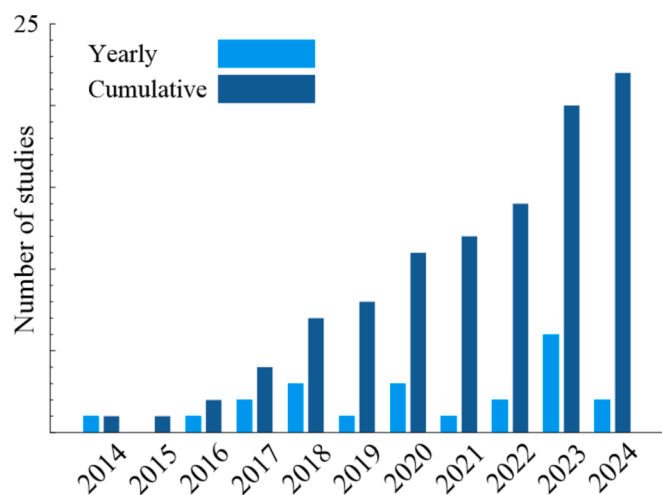


Fig. 4. Number of studies per year on concrete carbon mixing included in the systematic literature review.

explored this option. Lastly, the carbon dioxide injection system can be directly added to the mixer with a retrofitting operation. As a result, all the components of the mix are simultaneously amalgamated to form the carbonated concrete (Fig. 5c). The containers adopted for the mixing process in the second and third case can be either airtight to ensure that carbon dioxide is fixed into the cementitious matrix or eventually mixers with the open lid. The latter case benefits from its simplicity since the mixing operations can be conducted under atmospheric pressure. Each of the systems described has its own peculiarities that can strongly affect the outcome of the carbonation process, and they are therefore investigated separately.

The sample size is a critical factor for technological deployment, and therefore, it is important to highlight the systems used in each study. Of the 22 works included in the review, five implemented the injection system on an industrial scale, specifically using drum mixers or concrete trucks. Two studies developed a sealed mixer with a batching volume of approximately 20 L, effectively simulating conventional on-site mixers. In contrast, most studies produced pastes and mixes at the laboratory scale, which may present significant challenges when scaling up to industrial volumes.

### 3.3. Carbon dioxide source

The source of CO<sub>2</sub> plays a significant role for the development of an industrial process as well as in the environmental assessment. Hence, a separate remark is dedicated to the type of carbon dioxide used in the studies included in the review. Firstly, the use of pure CO<sub>2</sub> gas implies an upstream capture facility with a purification system (Aaron and Tsouris, 2005). Conversely, flue gases feature lower CO<sub>2</sub> concentrations (Feng et al., 2022) and the presence of pollutants that might impair the efficiency of the carbonation process (Xian et al., 2021). However, 23 of the 25 studies used pure CO<sub>2</sub> either in its gaseous form (18) or as liquified CO<sub>2</sub> (5). The use of liquified carbon dioxide implies the phase transition at the delivery into the mix, creating the so-called CO<sub>2</sub>-snow (Monkman et al., 2016). Only one study considered the use of diluted CO<sub>2</sub> gas including both an air tank and a CO<sub>2</sub> gas tank and a mixing system. The investigated concentrations of 50 % and 100 % aimed at mimicking the implications of an industrial-like flue gas in place of the pure carbon dioxide. The results showed a reduced temperature increment of the fresh concrete mixed with 50 % CO<sub>2</sub> compared to the pure gas. Nonetheless, the authors did not discuss the effects of the concentration on the mechanical performance. Moreover, the presence of pollutants was not considered for the low concentration case. Another study explored the use of dry ice in the mix instead of gaseous carbon dioxide (Chechani et al., 2023). The temperature change caused by the addition of the dry ice significantly reduced the setting time and slightly reduced the compressive strength with respect to the gaseous CO<sub>2</sub> mix. In addition, the enhancement of CaCO<sub>3</sub> formation due to the carbonation process was less relevant for the samples mixed with dry ice.

## 4. Results and discussion of the review

The 22 studies included in the review were separated in the three categories described in Section 3. Then, a comparison among the investigated works was conducted considering the following aspects.

- The type of cementitious material used for the sequestration.
- The process parameters, including pressure, temperature, and duration of the mixing phase.
- The efficiency of the sequestration process, with respect to the uptake of carbon dioxide.
- The effects of the enforced carbonation on the mechanical performance of the final product.
- The effects of the enforced carbonation on the durability of the final product.

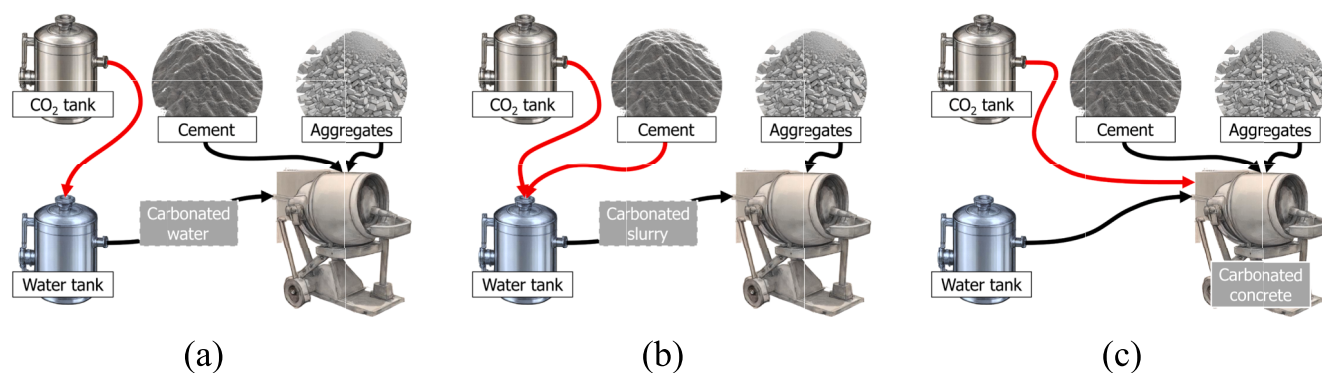


Fig. 5. Possible processes to produce ready-mix carbonated concrete: (a) carbonation of the mixing water, (b) carbonation of the cement slurry, and (c) direct injection during the mixing process.

#### 4.1. Carbonated water

The injection of carbon dioxide inside the mixing water does not directly trigger the carbonation since the reactive compounds are present in the constituents added afterwards. However, the underlying processes are the ones described in the introduction. In addition, the production line can be easily adapted by implementing an injection system for the carbon dioxide together with a sealed tank to carbonate the water, which is then discharged into the concrete mixer. This kind of approach was firstly proposed by Kwasny et al. (Kwasny et al., 2014), and five studies in total were attributed to this category.

A study by Qin et al. (Qin et al., 2018) investigated the use of a nano- $\text{CaCO}_3$  suspension as replacement of mixing water. The purpose of this approach is twofold; on the one hand, the calcium carbonate nanoparticles can be produced through carbonation, thus sequestering  $\text{CO}_2$  in the process (Han et al., 2023). In addition, the nanoparticles present in the mix can provide a nucleation effect to enhance the hydration and a filler effect to densify the matrix, thus promoting the strength development of the produced concrete (Fu et al., 2022). However, since the production of the nano- $\text{CaCO}_3$  suspension is a separate process, the study was excluded from the review.

Three relevant parameters were identified for the water carbonation process, including the pressure and time required to obtain a stable solution and the solution pH, the latter being particularly relevant with respect to the restrictions concerning the mixing water for concrete production. In fact, the pH of the mixing water can affect the mechanical, rheological, and durability properties of concrete due to the different alkalinity of the environment during hydration (Saha et al., 2022; Utepov et al., 2022). Therefore, National and International Standards prescribe minimum pH values among the chemical properties of mixing water (Table 2).

The carbonation of water is a slow and energy-intensive process that requires elevated pressures. The solubility is governed by Henry's Law, which yields a solubility of  $\text{CO}_2$  in water equal to  $3.3 \times 10^{-2}$  mol/L (1.45 g/L) at atmospheric pressure and 25 °C temperature. To enhance the solubility, lower temperatures and higher pressures are required. In addition, the time required for the solution to fully carbonated depends on the environmental conditions, and it can be estimated with Fick's Diffusion Law, where the diffusion coefficient varies with temperature. Other parameters affecting the diffusion rate are the stirring condition

and the  $\text{CO}_2$  concentration gradient. The minimum considered time among the studies was 24 h for carbonation at 40 bar pressure (Kwasny et al., 2014), although the additional time due to pressure decrease and compensation was not reported, whereas for an injection pressure of 8 bar, the time required was 3 days (Lippiatt and Ling, 2020; Lippiatt et al., 2019). In one case the carbonated water used was a commercially available sparkling water and thus the process parameters were not specified (Suescum-Morales et al., 2022; Suescum-Morales et al., 2022). Finally, Kim et al. (Kim et al., 2020) proposed an alternative and more efficient approach based on carbon dioxide nanobubbles generated by cavitation with a tailored equipment. Nanobubbles are characterized by a small buoyancy and therefore they can last longer in suspension (Cerrón-Calle et al., 2022), partially addressing the natural evaporation of regular bubbles that affect the quantity of  $\text{CO}_2$  effectively delivered into the concrete mix (Lippiatt and Ling, 2020; Lippiatt et al., 2019). Nevertheless, the carbon dioxide uptake due to the use of  $\text{CO}_2$  nanobubble water into the mix was not assessed. The lack of process parameters in the studies considered for the present section poses a strong limitation in the understanding of the effectiveness of using carbonated water for concrete mixing. Moreover, when the  $\text{CO}_2$  uptake is not directly assessed it is only possible to derive qualitative information on the properties of the carbonated concrete.

Table 3 provides the summary of the different approaches for concrete production with carbonated water. The carbonation process significantly reduces the pH of water, hence eventually not being compliant with the current Standards. Moreover, the time and pressure required to achieve a stable carbonated solution might not meet the continuous demand of concrete on site or in batching plants. Alternatively, the use of commercial sparkling water can be beneficial for concrete properties as well as the implementation of a nanobubble injector in the water tank (Suescum-Morales et al., 2022; Suescum-Morales et al., 2022; Kim et al., 2020). The presence of carbon dioxide in the suspension enabled the carbonation reactions that consequently enhanced the compressive strength of the final product. The  $\text{CO}_2$  uptake with these methods is limited if compared to direct injection systems due to the volatility of  $\text{CO}_2$  bubbles. Therefore, small amounts of sequestered  $\text{CO}_2$  promoted the strength development of the carbonated product. Lastly, high carbon dioxide uptake values led to a significant performance reduction since the heat generation during hydration was strongly affected using carbon dioxide in the mix (Kwasny et al., 2014).

Table 2

Minimum pH requirements for some International and National Standards.

| Standard         | Ref.                                                               | Context                  | Minimum pH |
|------------------|--------------------------------------------------------------------|--------------------------|------------|
| ISO 12439:2010   | (International Organization for Standardization, 2010)             | International            | 5.0        |
| UNI EN 1008:2003 | (Ente italiano di normazione, UNI EN, 1008)                        | National (Italy)         | 4.0        |
| JGJ 63:2006      | (Ministry of Construction of the people's Republic of China, 2006) | National (China)         | 4.5        |
| ASTM C1602:2022  | (ASTM International, 2022)                                         | National (United States) | n.d.       |

**Table 3**

Summary of the process parameters for water carbonation and the effects of carbonated water used as mixing water for concrete production.

| Study                                                        | Carbonation |      | Performance |                        |           |                    |                   |
|--------------------------------------------------------------|-------------|------|-------------|------------------------|-----------|--------------------|-------------------|
|                                                              | Pressure    | Time | pH          | CO <sub>2</sub> uptake | Heat flow | Compress. strength | Flexural strength |
| (Kwasny et al., 2014)                                        | 40 bar      | 1 d  | n.d.        | 3.9 %                  | ↑         | –                  | –                 |
| (Lippiatt and Ling, 2020; Lippiatt et al., 2019)             | 8 bar       | 3 d  | 4.2         | 0.6 %                  | ↑         | ↓                  | –                 |
| (Kim et al., 2020)                                           | Ambient     | n.d. | 4.5         | n.d.                   | –         | ↑                  | ↑                 |
| (van Tonder and Low, 2021)                                   | Ambient     | n.d. | n.d.        | n.d.                   | –         | ↑                  | –                 |
| (Suescum-Morales et al., 2022; Suescum-Morales et al., 2022) | n.d.        | n.d. | 4.8         | 0.8 %                  | –         | ↑                  | ↑                 |

↑ = Increase; ↓ = Decrease

It is however necessary to underline that in the study by Kwasny et al. (Kwasny et al., 2014) the negative effect was attributed to the dual carbonation conducted both through water carbonation and gaseous injection during the mixing stage. Therefore, the two processes shall be assessed separately to better understand the contribution of each carbonation system to the properties of the final product.

#### 4.2. Carbonation of the cement slurry

The second stage of concrete production is the mixing between the reactive binder – OPC and eventual SCMs – and the water to trigger the hydration reaction that develops the strength. Most of the studies conducted at laboratory scale investigated the injection of carbon dioxide in the cement paste. On the one hand, this approach is easier to implement at smaller scales. In addition, the injection into the slurry targets specifically the reactive cementitious feedstock, guaranteeing the most efficient carbonation. In fact, the reactive surface exposed to the gas is maximized during this stage. Nevertheless, hydration might be partially hindered by the simultaneous carbonation, thus limiting the hydration extent and, consequently, the strength development of the final product. This interplay is governed both by the injection parameters (i.e., quantity, time) as an input, and by the pH of the solution. Since CO<sub>2</sub> injection occurs within the first few minutes of mixing, there is limited formation of portlandite, as hydration is still in its early stages. Initially, the pH drops due to the formation of carbonate and bicarbonate ions (Back et al., 2011; Ho et al., 2020; Jiang et al., 2022), and it then rises slightly as calcium carbonate precipitates (Li et al., 2023; Fu et al., 2024). However, only a limited amount of clinker undergoes carbonation, and with appropriate control of injection timing and quantity, hydration can proceed after carbonation. This allows portlandite to form, which gradually buffers the pH to higher, stable levels. The competition between hydration and carbonation was extensively discussed in the literature with specific regards to the phase assemblage of the reaction products (Peter et al., 2008; Shah et al., 2019). Zajac et al. (Zajac et al., 2020) observed that the carbonation of a cement paste resulted in significant production volumes of C-S-H, characterized by lower Ca/Si and higher Al/Si, and hemi- and mono-carbonate, when compared to untreated Portland cement paste. Therefore, the system exhibited decreased porosity and pore refinement. Hu et al. (Hu et al., 2022) analyzed the phase composition of the carbonated paste via XRD. The prevailing CaCO<sub>3</sub> form observed was calcite, with no aragonite. In the initial stage (4 h), C<sub>2</sub>S and C<sub>3</sub>S were detected, albeit significantly reduced at 28 days of hydration age. Hence, normal hydration took place despite the initial carbonation. Additionally, the enforced carbonation of the paste is strongly influenced by the presence of alkalis, which varies with the cement type. This effect is particularly pronounced in the initial stages, where alkalis significantly accelerate the carbonation rate (Zajac et al., 2021; Zajac et al., 2021).

Table 4 summarizes the studies that implemented the carbon dioxide injection on the slurry or paste. The subsequent addition of the other ingredients (e.g., aggregates) was not considered as minimum requirement for the inclusion in the present category. All the processes were performed in a sealed mixer without externally imposed pressure. In addition, in all cases the injection was executed at ambient temperature. Therefore, the two process parameters considered for the comparison

were the gas flow, where reported, and the mixing time. The former was generally controlled by a flowmeter valve placed between the carbon dioxide gas cylinder and the sealed mixer. Thus, it was possible to govern the dosage of CO<sub>2</sub> present in the environment. Nonetheless, in some cases the absorption of CO<sub>2</sub> was lower than the injected amount due to uncomplete reaction (Qin et al., 2018). The gas flow showed significant variability among the studies that reported its value and there is no evident correlation with the carbon dioxide uptake. Conversely, the mixing time, and particularly the carbon dioxide mixing time, directly affected the efficiency of the process. Specifically, Zhang et al. (Zhang et al., 2024) assessed the correlation between the mixing protocol and the carbon dioxide uptake. Longer injection times were associated with higher CO<sub>2</sub> uptake values, although the difference between a fast initial injection (90 s) and significantly longer times (>10 min) was comparatively modest. Additionally, the other study with long injection time (10 min) achieved the highest uptake value among the results considered (5 %) (Chechani et al., 2023). It is necessary to underline the need for direct measurements of the CO<sub>2</sub> uptake, which can strongly differ from the injected quantity due to system losses and limited absorption capacity of the feedstock. Hence, since most studies only reported dosed quantities of the carbon dioxide, analytical measurements are required to reinforce the correlation between the results and the input process parameters.

The compressive strength at 28 days was used as benchmark to evaluate the effectiveness of the carbon mixing process since it was the only parameter available in all the studies considered. The results obtained by He et al. (He et al., 2017) were neglected due to the remarkable differences with respect to all the other studies. The negative effects of the carbonation mixing measured in this study were attributed to the clusters formed by the particles during the microstructure development that created a porous structure. These clusters were then enclosed in a dense coating of carbonation products that prevented further hydration, thus hindering the strength of the material. Moreover, the consumption of calcium ions reduced the Ca/Si ratio of the C-S-H gel, hence decreasing its density (Suda et al., 2015; Königsberger et al., 2016). Zhang et al. (Zhang et al., 2024) obtained a similar outcome for the longest carbonation mixing, suggesting that excessive carbonation might negatively affect the performance of the final product. The reduction was attributed to the protective shell formed by the carbonation products around the unhydrated cement particles.

The results reported in Fig. 6 show the effects of concrete carbon mixing on the compressive strength of the final cementitious product. Only one of the cases considered experienced a reduction in the performance due to the higher CO<sub>2</sub> uptake compared to the other results from the same study (Zhang et al., 2024). To determine a correlation between CO<sub>2</sub> uptake and mechanical performance, three experimental data points were excluded from the analysis due to their notable deviation from the rest of the results. Even so, the reliability of the established correlation was limited by the scattering among the experimental results. In most cases the carbon dioxide uptake stands below 2 % and the variation of strength is not consistent with the input values. Therefore, although the carbonation of the cement pastes proved the beneficial contribution provided by the CO<sub>2</sub> injection in the fresh state, the reliability and consistency of the process require further investigation.



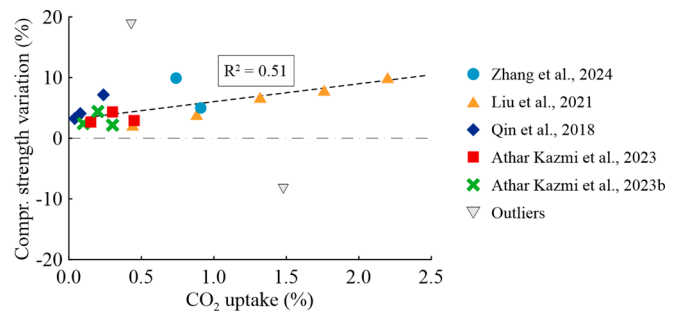
**Table 4**  
Summary of the process parameters for the carbonation of cement paste and the resulting carbon dioxide uptake and compressive strength variation.

| Study                                                                         | Process parameters                            |           |                                 | CO <sub>2</sub> uptake | Compr. strength <sup>(a)</sup> |
|-------------------------------------------------------------------------------|-----------------------------------------------|-----------|---------------------------------|------------------------|--------------------------------|
|                                                                               | Protocol                                      | Gas flow  | Mixing time                     |                        |                                |
| (Athar Kazmi and L. Vara Prasad Meesaraganda, P. Suresh Chandra Babu, , 2023) | Injection in the cement paste                 | n.d.      | 60 s                            | 0.15 %                 | +2.67 %                        |
|                                                                               |                                               |           |                                 | 0.30 %                 | +4.31 %                        |
|                                                                               |                                               |           |                                 | 0.45 %                 | +2.91 %                        |
| (Athar Kazmi and Prasad, 2023)                                                | Injection in the cement slurry                | n.d.      | 60 s                            | 0.10 %                 | +2.37 %                        |
|                                                                               |                                               |           |                                 | 0.20 %                 | +4.37 %                        |
|                                                                               |                                               |           |                                 | 0.30 %                 | +2.19 %                        |
| (Chechani et al., 2023)                                                       | Injection in the cement paste                 | 1.5 bar   | 5 + 10 min                      | 5 %                    | +9%                            |
| (He et al., 2017)                                                             | Injection in the cement paste                 | 5 L/ min  | 240 s                           | 3.4 %                  | -83 %                          |
|                                                                               | Injection in the cement paste + air mixing    |           |                                 | 240 + 120 s            | 10.1 % <sup>(b)</sup>          |
| (Liu et al., 2021)                                                            | Injection in the cement paste                 | n.d.      | <sup>(c)</sup>                  | 0.44 %                 | +2.13 %                        |
|                                                                               |                                               |           |                                 | 0.88 %                 | +3.87 %                        |
|                                                                               |                                               |           |                                 | 1.32 %                 | +6.70 %                        |
|                                                                               |                                               |           |                                 | 1.76 %                 | +7.95 %                        |
|                                                                               |                                               |           |                                 | 2.20 %                 | +10.03 %                       |
| (Qian et al., 2018)                                                           | Pre-carbonation of slaked lime slurry         | 2 L/ min  | 10 min                          | <sup>(d)</sup>         | +16 %                          |
| (Qin et al., 2018)                                                            | Pre-carbonation of Ca(OH) <sub>2</sub> slurry | 20 L/ min | 15 min                          | <sup>(d)</sup>         | +13 %                          |
|                                                                               |                                               |           | 150 s/ 1 <sup>(e)</sup> + 330 s | 0.04 %                 | +3.27 %                        |
|                                                                               |                                               |           | 0.08 %                          | +4.08 %                |                                |
| (Zhang et al., 2024)                                                          | Injection in the cement paste                 | n.d.      | 90 s + 60 min                   | 0.43 %                 | +18.76 %                       |
|                                                                               |                                               |           | 10 + 10 + 40 min                | 0.74 %                 | +9.91 %                        |
|                                                                               |                                               |           | 20 + 40 min                     | 0.91 %                 | +4.96 %                        |
|                                                                               |                                               |           | 60 min                          | 1.48 %                 | -8.32 %                        |
|                                                                               |                                               |           | 0.24 %                          | +7.19 %                |                                |

**Bold** = CO<sub>2</sub> mixing; *Italics* = Air mixing  
 (a) Variation with respect to non-carbonated samples (28 d); (b) includes CO<sub>2</sub> curing;  
 (c) upon reaching the prescribed absorption; (d) 1 % and 3 % replacement of cement with slaked lime; (e) per liter of solution;  
 \*dosed, not measured

4.3. Concrete mixing injection

The direct injection of gaseous carbon dioxide into fresh concrete was investigated in the recent years by various authors, albeit being already industrialized by the company CarbonCure Technologies Inc. that developed an injection system to sequester CO<sub>2</sub> during the mixing in the trucks (Monkman and MacDonald, 2017). The primary goal of the



**Fig. 6.** Correlation between the CO<sub>2</sub> uptake and the variation of the compressive strength with respect to the non-carbonated sample after 28 days of curing.

research was to enhance the uptake capacity of carbonated concrete while simultaneously maintaining all its material properties. To do so, the process parameters were varied among the studies, with particular emphasis towards the mixing procedure (Li et al., 2023), the CO<sub>2</sub> dosage (Monkman et al., 2016; Rashid and Singh, 2023; Monkman et al., 2023), the type of binder and water-to-binder ratio (van Tonder and Low, 2021; Rashid and Singh, 2023; Lee et al., 2018), and the delivery pressure (Lee et al., 2018). The performance assessment of the carbonated concrete is herein divided into three main categories, namely the fresh properties, the mechanical performance, and the durability.

4.3.1. Fresh state properties

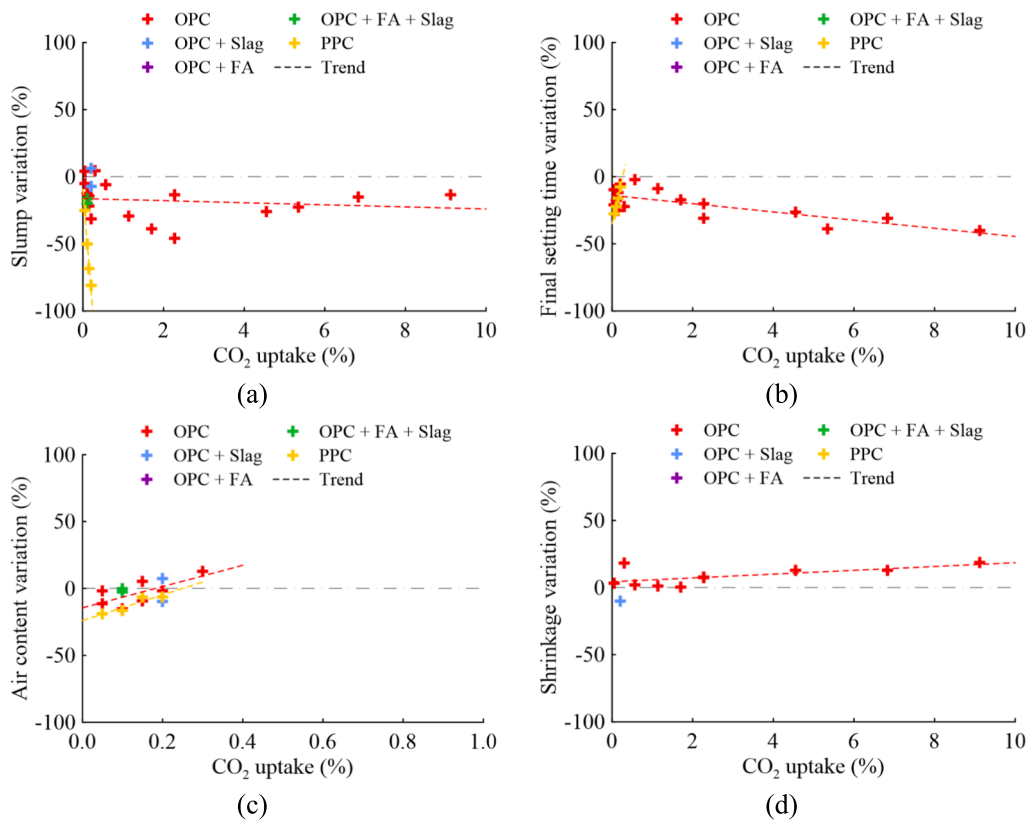
Among the properties of concrete in its fresh state, a major concern for carbonated concrete revolves around the accelerating effects on the hardening process due to CO<sub>2</sub> injection. Therefore, the change in the final setting time and the flowability of carbonated concrete were assessed by most authors. Table 5 reports the summary of the results available in literature, emphasizing the extent to which early age carbonation hindered the workability of concrete, accelerated its setting time, interacted with the air voids development, and increased the drying shrinkage (Fig. 7). The results are consistent across the studies reviewed in the literature. However, a few cases reported opposite effects, particularly Monkman et al. (Monkman et al., 2016) and Monkman et al. (Monkman et al., 2023). These studies observed both an increase in the slump of fresh concrete and a higher air content (Fig. 7a and Fig. 7c). No explanation was provided for this phenomenon, which may be attributed to variability in the results. Higher CO<sub>2</sub> dosages further reduced the workability, as in the work by Rashid and Singh (Rashid and Singh, 2023), while Li et al. (Li et al., 2023) reported a threshold of mixing time beyond which the negative effects were partially recovered (Fig. 7a). In addition, the secondary air mixing after injection significantly mitigated the slump reduction and the acceleration of the final setting time, while the shrinkage remained unaffected. The type of binder also influenced the workability of concrete. The pozzolana cement used by Rashid and Singh (Rashid and Singh, 2023) and the blended cement with fly ash and slag by Monkman and MacDonald (Monkman and MacDonald, 2017) experienced significant slump reduction even at small CO<sub>2</sub> dosages (Fig. 7a).

4.3.2. Mechanical performance

The mechanical performance of carbonated concrete is generally referred to as the benchmark to evaluate the effectiveness of the injection process. Therefore, all the surveyed studies assessed the compressive strength of carbonated concrete with respect to its reference. Table 6 reports the variation in the compressive strength due to the carbonation. In 22 of the 38 cases, the injection of carbon dioxide enhanced the mechanical performance of the material (Fig. 8). However, in most instances, this effect was reversed when higher CO<sub>2</sub> dosages were introduced into the mixer. In fact, excessive CO<sub>2</sub> dosages might be detrimental to the hydration reaction and, hence, the

**Table 5**  
Variation in concrete fresh properties due to carbon dioxide injection.

| Study                         | Carbonation        |                                                 |                        | Properties |           |             |           |
|-------------------------------|--------------------|-------------------------------------------------|------------------------|------------|-----------|-------------|-----------|
|                               | Binder (w/b)       | Process details                                 | CO <sub>2</sub> dosage | Slump      | Final set | Air content | Shrinkage |
| (Rashid and Singh, 2023)      | OPC (0.45)         | Injection of 20 bar pressurized CO <sub>2</sub> | 0.05 %                 | -5.0 %     | -9.7 %    | -11.3 %     |           |
|                               |                    |                                                 | 0.10 %                 | -12.5 %    | -17.8 %   | -15.1 %     |           |
|                               |                    |                                                 | 0.15 %                 | -21.9 %    | -12.0 %   | -9.4 %      |           |
|                               |                    |                                                 | 0.20 %                 | -31.3 %    | -5.7 %    | -1.9 %      |           |
|                               |                    |                                                 | 0.05 %                 | -25.0 %    | -27.5 %   | -18.8 %     |           |
| (Li et al., 2023)             | OPC (0.35)         | Mixing: Air 240 s + CO <sub>2</sub> 15 s        | 0.10 %                 | -50.0 %    | -22.5 %   | -16.7 %     |           |
|                               |                    |                                                 | 0.15 %                 | -68.8 %    | -15.8 %   | -6.3 %      |           |
|                               |                    |                                                 | 0.20 %                 | -81.3 %    | -7.5 %    | -6.3 %      |           |
|                               |                    |                                                 | 0.56 %                 | -6.2 %     | -2.2 %    |             | +1.8 %    |
|                               |                    |                                                 | 1.13 %                 | -29.2 %    | -8.9 %    |             | +0.9 %    |
|                               |                    |                                                 | 1.71 %                 | -39.0 %    | -17.4 %   |             | +0.2 %    |
|                               |                    |                                                 | 2.28 %                 | -46.2 %    | -31.1 %   |             | +7.5 %    |
|                               |                    |                                                 | 4.55 %                 | -26.2 %    | -26.7 %   |             | +12.9 %   |
|                               |                    |                                                 | 6.83 %                 | -15.4 %    | -31.1 %   |             | +12.7 %   |
| (Saikia and Rajput, 2024)     | OPC (0.45)         | Mixing: Air 240 s + CO <sub>2</sub> 240 s       | 9.12 %                 | -13.3 %    | -40.0 %   |             | +18.5 %   |
|                               |                    |                                                 | 2.28 %                 | -13.3 %    | -20.4 %   |             | +7.9 %    |
| (Monkman et al., 2016)        | OPC (0.39)         | Injection in truck load                         | 5.35 %                 | -22.7 %    | -38.9 %   |             |           |
|                               |                    |                                                 | 0.05 %                 | +4.0 %     | -20.8 %   | -2.0 %      | +3.0 %    |
|                               |                    |                                                 | 0.15 %                 | -14.3 %    | -25.3 %   | +5.1 %      |           |
| (Monkman and MacDonald, 2017) | OPC-FA-Slag (0.34) | Injection in the center of the mixer            | 0.30 %                 | +4.5 %     | -22.3 %   | +12.7 %     | +18.2 %   |
|                               |                    |                                                 | 0.10 %                 | -19.7 %    |           | 0.0 %       |           |
|                               |                    |                                                 | 0.10 %                 | -16.4 %    |           | -2.9 %      |           |
| (Monkman et al., 2023)        | OPC-Slag Mix A     | Injection in the batching streamline            | 0.20 %                 | -7.2 %     |           | +7.5 %      | -10.0 %   |
|                               |                    |                                                 | 0.20 %                 | +6.2 %     |           | -9.9 %      |           |



**Fig. 7.** Correlation between the CO<sub>2</sub> uptake and the variation of the fresh properties of carbonated concrete based on the type of binder; (a) slump (Li et al., 2023; Saikia and Rajput, 2024; Monkman et al., 2016; Monkman and MacDonald, 2017; Rashid and Singh, 2023; Monkman et al., 2023), (b) final setting time (Li et al., 2023; Saikia and Rajput, 2024; Monkman et al., 2016; Rashid and Singh, 2023), (c) air content (Monkman et al., 2016; Monkman and MacDonald, 2017; Rashid and Singh, 2023; Monkman et al., 2023), and (d) shrinkage (Li et al., 2023; Monkman et al., 2016; Monkman et al., 2023).

development of concrete microstructure. The partial replacement of OPC with alternative binders, such as pozzolana Portland cement (PPC) (Rashid and Singh, 2023) or fly ash (van Tonder and Low, 2021), influenced the effects of CO<sub>2</sub> injection on compressive strength, either

by further reducing the mechanical performance or by partly mitigating the beneficial effects, respectively (Fig. 8a). Furthermore, the application of additional pressure was found to be detrimental to the material performance, while its contribution to the uptake remained

**Table 6**  
Variation in concrete compressive strength due to carbon dioxide injection.

| Study                         | Binder (w/ b)      | Process details                                      | CO <sub>2</sub> dosage | Compr. strength |
|-------------------------------|--------------------|------------------------------------------------------|------------------------|-----------------|
| (Rashid and Singh, 2023)      | OPC (0.45)         | Injection of 20 bar pressurized CO <sub>2</sub>      | 0.05 %                 | +3.6 %          |
|                               |                    |                                                      | 0.10 %                 | +9.6 %          |
|                               |                    |                                                      | 0.15 %                 | -2.0 %          |
|                               |                    |                                                      | 0.20 %                 | -7.0 %          |
|                               |                    |                                                      | 0.05 %                 | +19.3 %         |
| (van Tonder and Low, 2021)    | OPC (0.67)         | Injection in the formwork during mixing              | n.d.                   | +12.8 %         |
|                               |                    |                                                      | n.d.                   | +12.8 %         |
|                               |                    |                                                      | n.d.                   | +14.1 %         |
|                               |                    |                                                      | n.d.                   | +12.0 %         |
|                               |                    |                                                      | n.d.                   | +17.8 %         |
| (Lee et al., 2018)            | OPC (0.75)         | Injection in pressurized mixer (2 bar)               | ≤ 9.5 %                | -14.3 %         |
|                               |                    | Injection in pressurized mixer (4 bar)               | ≤ 9.5 %                | -12.3 %         |
|                               |                    | Injection in pressurized mixer (6 bar)               | ≤ 9.5 %                | -22.4 %         |
|                               | OPC (0.65)         | Injection in pressurized mixer (2 bar)               | ≤ 9.5 %                | -20.7 %         |
|                               |                    | Injection in pressurized mixer (4 bar)               | ≤ 9.5 %                | -18.3 %         |
|                               |                    | Injection in pressurized mixer (6 bar)               | ≤ 9.5 %                | -22.7 %         |
|                               | OPC (0.55)         | Injection in pressurized mixer (2 bar)               | ≤ 9.5 %                | -33.5 %         |
|                               |                    | Injection in pressurized mixer (4 bar)               | ≤ 9.5 %                | -27.3 %         |
|                               |                    | Injection in pressurized mixer (6 bar)               | ≤ 9.5 %                | -38.8 %         |
|                               |                    | Injection in pressurized mixer (6 bar)               | ≤ 9.5 %                | -38.8 %         |
| (Li et al., 2023)             | OPC (0.35)         | Mixing: Air 240 s + CO <sub>2</sub> 15 s             | 0.56 %                 | +4.4 %          |
|                               |                    | Mixing: Air 240 s + CO <sub>2</sub> 30 s             | 1.13 %                 | +21.0 %         |
|                               |                    | Mixing: Air 240 s + CO <sub>2</sub> 45 s             | 1.71 %                 | -2.0 %          |
|                               |                    | Mixing: Air 240 s + CO <sub>2</sub> 60 s             | 2.28 %                 | +1.9 %          |
|                               |                    | Mixing: Air 240 s + CO <sub>2</sub> 100 s            | 4.55 %                 | +23.8 %         |
|                               |                    | Mixing: Air 240 s + CO <sub>2</sub> 150 s            | 6.83 %                 | +23.6 %         |
|                               |                    | Mixing: Air 240 s + CO <sub>2</sub> 240 s            | 9.12 %                 | +28.0 %         |
|                               |                    | Mixing: Air 240 s + CO <sub>2</sub> 60 s + Air 180 s | 2.28 %                 | +31.1 %         |
| (Saikia and Rajput, 2024)     | OPC (0.45)         | Mixing: CO <sub>2</sub> 240 s + Air 60 s             | 5.35 %                 | +2.9 %          |
| (Monkman et al., 2016)        | OPC (0.39)         | Injection in truck load                              | 0.05 %                 | +3.0 %          |
|                               |                    |                                                      | 0.15 %                 | -4.0 %          |
|                               |                    |                                                      | 0.30 %                 | -6.0 %          |
| (Monkman and MacDonald, 2017) | OPC-FA-Slag (0.34) | Injection in the center of the mixer                 | 0.10 %                 | +15.4 %         |

**Table 6 (continued)**

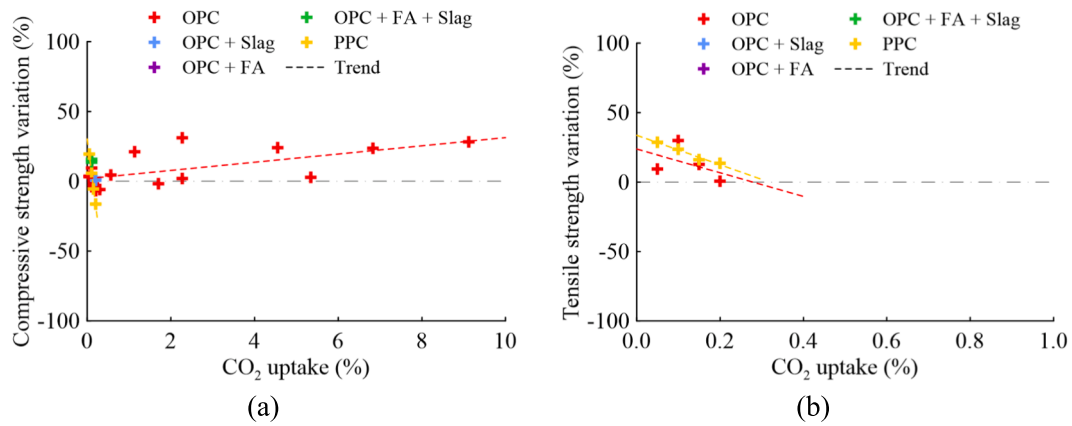
| Study                  | Binder (w/ b)      | Process details                      | CO <sub>2</sub> dosage | Compr. strength |
|------------------------|--------------------|--------------------------------------|------------------------|-----------------|
| (Monkman et al., 2023) | OPC-FA-Slag (0.35) | Injection in the center of the mixer | 0.10 %                 | +13.7 %         |
|                        |                    | Injection in the batching streamline | 0.20 %                 | +0.7 %          |
|                        |                    | Injection in the batching streamline | 0.20 %                 | +2.2 %          |
|                        |                    | Injection in the batching streamline | 0.20 %                 | +2.2 %          |

unconfirmed due to a lack of measurements (Lee et al., 2018). The microstructure development benefited from the secondary air mixing after the carbon dioxide injection, as observed by Li et al. (Li et al., 2023). The increase in compressive strength was higher than the other investigated cases despite the limited amount of CO<sub>2</sub> with respect to longer carbon dioxide mixing durations. This result may confirm the description of the microstructure development proposed by He et al. (He et al., 2017). In fact, the coating effect due to the layer of carbonates precipitating after early carbonation might be effectively prevented with further mixing. Hence, hydration can progress while benefiting from the precipitation of nano-CaCO<sub>3</sub> (Zhang et al., 2018).

Finally, the mechanical performance of carbonated concrete requires broader investigation also encompassing tensile strength and fracture toughness, which are crucial to the durability of reinforced concrete elements. Currently, there is a lack of data beyond the compressive strength, as remarked by Lim et al. (Lim et al., 2019). Among the reviewed studies, only one assessed the impact of carbon dioxide injection on tensile strength (Rashid and Singh, 2023), albeit across various scenarios (Table 7). Both OPC and PPC concrete remarkably benefited from the addition of carbon dioxide (Fig. 8b). The highest increase in the tensile strength for OPC occurred with a CO<sub>2</sub> dosage of 0.10 %, with diminishing returns observed at higher doses. PPC exhibited a comparable trend, maintaining enhancements exceeding 13 % across all tested doses, albeit with a similar reduction in effectiveness at higher levels. The small dosage of CO<sub>2</sub>, however, implies that the correlation might not apply when increasing the injected gas, and the effects on the tensile strength shall be further investigated. Similarly, flexural strength was only evaluated in a singular instance by Monkman et al. (Monkman et al., 2023), who observed a marginal increase in performance by 1.3 %. However, given the minimal variation and the absence of comprehensive parametric investigations, drawing general conclusions regarding the effect of CO<sub>2</sub> injection on flexural strength is precluded.

#### 4.3.3. Durability

Concrete durability is a key issue in a performance-based design and assessment of reinforced concrete structures, mainly related to the protection that concrete provides to the embedded steel bars, which must preserve their tensile strength and ductility along the life cycle of the structure (Ahmad, 2003). The assessment of durability is a multi-faceted discipline that requires broad investigation with multiple tests. However, a major concern for concrete subject to early carbonation is the alkalinity of the solution, which must provide the passivation effect that prevents steel reinforcement corrosion (Bertolini et al., 2013). As reported by Berkeley and Pathmanaban (Berkeley and Pathmanaban, 1990), the threshold pH to trigger steel corrosion is 9.5. It is therefore necessary to determine the alkalinity of concrete after CO<sub>2</sub> injection, since calcium hydroxide consumption reduces concrete pH. Table 8 reports the pH values of different concrete mixes before and after the carbon dioxide injection treatment. The differences in the pH were negligible in all cases owing to the limited extent of carbonation due to the small CO<sub>2</sub> dosage as well as the progress of subsequent hydration that partially restored the alkalinity of the solution (Fig. 9a). This phenomenon was already observed for concrete carbonation curing by Zhang and Shao (Zhang and Shao, 2016) and Wei et al. (Wei et al., 2023). In addition, the densification obtained through early carbonation



**Fig. 8.** Correlation between the CO<sub>2</sub> uptake and the variation of the mechanical performance of carbonated concrete based on the type of binder; (a) compressive strength (Li et al., 2023; Saikia and Rajput, 2024; Monkman et al., 2016; Monkman and MacDonald, 2017; Rashid and Singh, 2023; Monkman et al., 2023), and (b) tensile strength (Rashid and Singh, 2023).

**Table 7**

Variation in concrete tensile strength due to carbon dioxide injection (Rashid and Singh, 2023).

| Binder (w/b) | Process details                                 | CO <sub>2</sub> dosage | Tensile strength |
|--------------|-------------------------------------------------|------------------------|------------------|
| OPC (0.45)   | Injection of 20 bar pressurized CO <sub>2</sub> | 0.05 %                 | +9.2 %           |
|              |                                                 | 0.10 %                 | +29.7 %          |
|              |                                                 | 0.15 %                 | +12.9 %          |
|              |                                                 | 0.20 %                 | +0.7 %           |
| PPC (0.45)   | Injection of 20 bar pressurized CO <sub>2</sub> | 0.05 %                 | +28.6 %          |
|              |                                                 | 0.10 %                 | +23.6 %          |
|              |                                                 | 0.15 %                 | +16.2 %          |
|              |                                                 | 0.20 %                 | +13.5 %          |

may enhance the resistance towards weathering carbonation by limiting the penetration of ions (Zhang and Shao, 2016; Xian et al., 2022).

Another major concern for reinforced concrete structures is the ingress of chloride ions, which may trigger the corrosion of the steel bars (Basheer et al., 2001). Therefore, the assessment of chloride penetration

**Table 8**

Variation in concrete durability properties due to carbon dioxide injection.

| Study                    | Carbonation  |                                                      |                        | Properties                               |                    |                         |         |  |        |
|--------------------------|--------------|------------------------------------------------------|------------------------|------------------------------------------|--------------------|-------------------------|---------|--|--------|
|                          | Binder (w/b) | Process details                                      | CO <sub>2</sub> dosage | Ref. pH                                  | CO <sub>2</sub> pH | RCPT                    | Voids   |  |        |
| (Rashid and Singh, 2023) | OPC (0.45)   | Injection of 20 bar pressurized CO <sub>2</sub>      | 0.05 %                 | 12.36                                    | 12.39              | -9.7 %                  | +2.0 %  |  |        |
|                          |              |                                                      | 0.10 %                 | 12.36                                    | 12.36              | -21.3 %                 | -8.0 %  |  |        |
|                          |              |                                                      | 0.15 %                 | 12.36                                    | 12.38              | -13.4 %                 | -1.0 %  |  |        |
|                          |              |                                                      | 0.20 %                 | 12.36                                    | 12.36              | -5.8 %                  | +16.5 % |  |        |
|                          | PPC (0.45)   | Injection of 20 bar pressurized CO <sub>2</sub>      | 0.05 %                 | 12.19                                    | 12.26              | -11.6 %                 | -5.4 %  |  |        |
|                          |              |                                                      | 0.10 %                 | 12.19                                    | 12.28              | +18.1 %                 | +8.8 %  |  |        |
|                          |              |                                                      | 0.15 %                 | 12.19                                    | 12.29              | +41.2 %                 | +11.8 % |  |        |
| (Li et al., 2023)        | OPC (0.35)   | Mixing: Air 240 s + CO <sub>2</sub> 15 s             | 0.56 %                 | 12.44                                    | 12.21              |                         |         |  |        |
|                          |              | Mixing: Air 240 s + CO <sub>2</sub> 30 s             | 1.13 %                 | 12.44                                    | 12.21              |                         | -17.7 % |  |        |
|                          |              | Mixing: Air 240 s + CO <sub>2</sub> 45 s             | 1.71 %                 | 12.44                                    | 12.21              |                         |         |  |        |
|                          |              | Mixing: Air 240 s + CO <sub>2</sub> 60 s             | 2.28 %                 | 12.44                                    | 12.21              |                         | -25.5 % |  |        |
|                          |              | Mixing: Air 240 s + CO <sub>2</sub> 100 s            | 4.55 %                 | 12.44                                    | 12.21              |                         | -26.6 % |  |        |
|                          |              | Mixing: Air 240 s + CO <sub>2</sub> 150 s            | 6.83 %                 | 12.44                                    | 12.21              |                         |         |  |        |
|                          |              | Mixing: Air 240 s + CO <sub>2</sub> 240 s            | 9.12 %                 | 12.44                                    | 12.21              |                         | -22.4 % |  |        |
|                          |              | Mixing: Air 240 s + CO <sub>2</sub> 60 s + Air 180 s | 2.28 %                 | 12.44                                    | 12.21              |                         | -26.0 % |  |        |
|                          |              | (Saikia and Rajput, 2024)                            | OPC (0.45)             | Mixing: CO <sub>2</sub> 240 s + Air 60 s | 5.35 %             | 13.00                   | 12.00   |  | -8.8 % |
|                          |              |                                                      |                        | (Monkman et al., 2016)                   | OPC (0.39)         | Injection in truck load | 0.05 %  |  |        |
| 0.15 %                   |              |                                                      | +2.2 %                 |                                          |                    | +24.5 %                 |         |  |        |
| 0.30 %                   |              |                                                      | -3.6 %                 |                                          |                    | -6.1 %                  |         |  |        |
| (Monkman et al., 2023)   | OPC-FA Mix A | Injection in the batching streamline                 | 0.10 %                 | 13.20                                    | 13.20              | -6.3 %                  |         |  |        |
|                          | OPC-FA Mix B | Injection in the batching streamline                 | 0.20 %                 | 13.20                                    | 13.10              | -8.3 %                  |         |  |        |

is of the utmost importance to understanding the durability of carbonated concrete. As shown in Table 8, the penetration of chlorides, evaluated using the rapid chloride penetration test (RCPT), was reduced in most cases due to the densification of the matrix (Fig. 9b). Conversely, for pozzolana Portland cement, an opposite effect was observed (Rashid and Singh, 2023), as the presence of finer particles diminished the densification effect induced by CO<sub>2</sub> injection. Moreover, carbonation processes are concurrent with pozzolanic reactions (Keppert et al., 2020). The same results were confirmed by the measurements of the volume of voids on the hardened concrete (Fig. 9c). In the study by Rashid and Singh (Rashid and Singh, 2023) the highest CO<sub>2</sub> dosage hindered the microstructure development, thus increasing the voids. Conversely, for lower CO<sub>2</sub> dosages the injection of carbon dioxide produced a dense matrix also in the hardened state, as confirmed by Li et al. (Li et al., 2023) and Monkman et al. (Monkman et al., 2016).

The overall effect of small dosages of carbon dioxide injection improved the durability of the concrete. However, higher dosages may negatively impact the hydration processes, potentially compromising the quality of the final product. Therefore, it is crucial to establish a proper balance between the need for increased CO<sub>2</sub> sequestration and concrete durability. Current literature, however, does not provide a



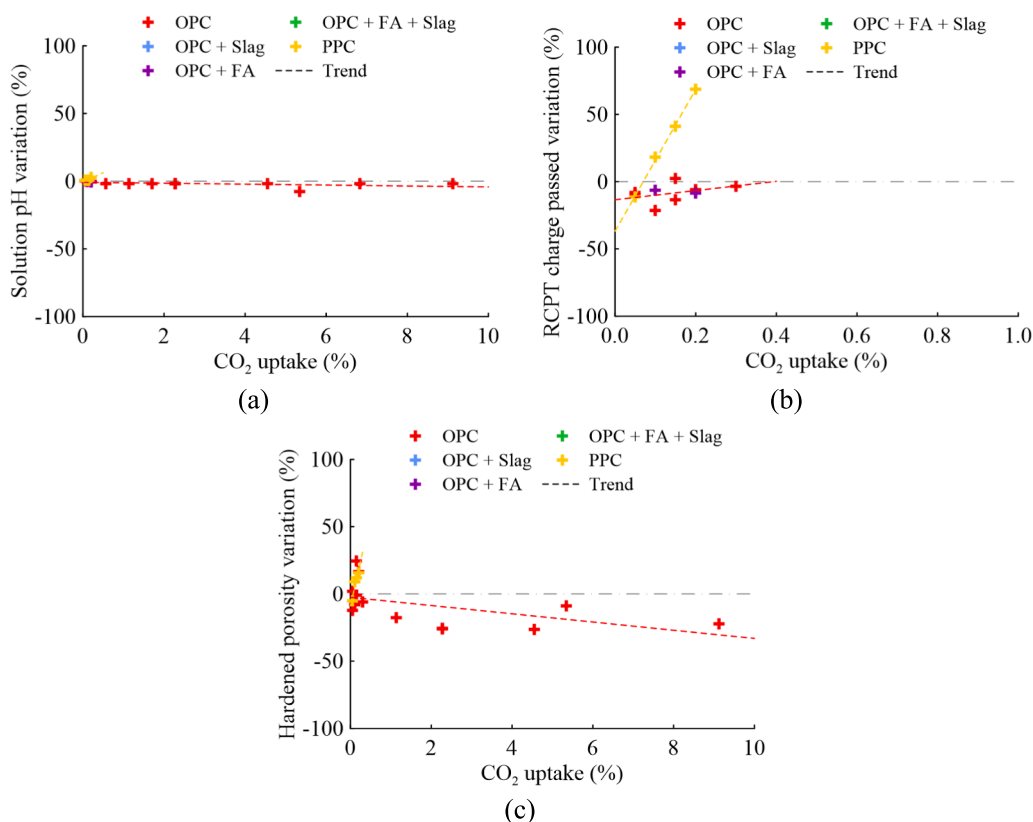


Fig. 9. Correlation between the CO<sub>2</sub> uptake and the variation of the durability properties of carbonated concrete based on the type of binder; (a) solution pH (Li et al., 2023; Saikia and Rajput, 2024; Rashid and Singh, 2023; Monkman et al., 2023), (b) RCPT charge passed (Monkman et al., 2016; Rashid and Singh, 2023; Monkman et al., 2023), and (c) porosity in the hardened state (Li et al., 2023; Saikia and Rajput, 2024; Monkman et al., 2016; Rashid and Singh, 2023).

clear threshold for adequate injection.

### 5. Environmental assessment

The availability of data on the processes involved for concrete carbon mixing is scarce due to the novelty of this technology. The implementation of environmental assessments is however necessary to understand the benefits of carbon dioxide injection for industrial concrete production. The present section addresses the environmental performance of the technology and its potential to curb the emissions of the concrete sector.

The life cycle assessment (LCA) is currently among the most systematic environment-oriented procedure for the sustainability assessment of products and industrial processes. None of the four studies included in the review specified that the environmental assessment conducted was an LCA in compliance with international guidelines ([139;140]). Despite the lack of standardization, the methodological approaches were deduced from the manuscripts, and the differences among the reviewed studies are herein discussed. The following aspects were considered for the comparison:

- Goal of the study;
- Assessment method;
- System boundaries;
- Functional unit;
- Impact categories.

Other relevant aspects of LCA, such as the handling of multifunctionality, temporal and geographical reference, data sources, and the handling of uncertainty were excluded from the review due to the lack of information in the available studies.

#### 5.1. Goal and scope

The goal of the studies was in all cases to evaluate a carbonated concrete mix against its baseline equivalent, hence the LCA qualifies as comparative assessment. The system boundaries were not specifically defined in any case with specific terms. It was however possible to infer them from the list of products and processes included in the calculation

Table 9

Summary of the processes included in the system boundaries for the studies considered.

| Process                                         | (Monkman and MacDonal, 2016) | (Monkman and MacDonal, 2017) | (Monkman, 2018) | (Zhang et al., 2024) |
|-------------------------------------------------|------------------------------|------------------------------|-----------------|----------------------|
| Production of concrete                          |                              | ●                            |                 | ●                    |
| CO <sub>2</sub> gas – Capture                   | ●                            | ●                            | ●               | ●                    |
| CO <sub>2</sub> gas – Transportation            | ●                            | ●                            | ●               | ●                    |
| Injection equipment – Production                |                              | ●                            | ●               |                      |
| Injection equipment – Transportation            |                              | ●                            | ●               |                      |
| Injection equipment – Operation                 |                              |                              | ●               | ●                    |
| CO <sub>2</sub> sequestration                   | ●                            | ●                            | ●               | ●                    |
| CO <sub>2</sub> savings due to mix optimization |                              | ●                            | ●               |                      |

(Table 9). As a result, the system boundaries were all categorized as cradle-to-gate. One of the studies referred to the Environmental Product Declarations (EPDs) and thus the details of the LCA methodology were not explicitly reported.

The cradle-to-gate boundary is a suitable approach to compare the production of concrete with carbon dioxide injection to its baseline equivalent. The modifications of the industrial equipment must be included to account for the additional impacts associated with the production, the transportation to the production site, and the operational consumptions. The first two categories require the estimation of the lifetime of the equipment, including eventual maintenance operations, since the additional impacts must be spread over the total production that the equipment can support. It is important to highlight that changes in the concrete microstructure due to CO<sub>2</sub> injection significantly impact the material's durability. In many cases, the refined pore structure resulting from accelerated carbonation enhances durability, as discussed in Section 4.3.3. Therefore, a cradle-to-grave analysis should be conducted to assess the full benefits of CO<sub>2</sub> injection across the concrete's service life. This analysis should also consider the balance between early-stage GHG sequestration and the reduced carbonation potential during service life, which may limit additional CO<sub>2</sub> uptake. Over the long term, this could mean that enforced carbonation may not yield net benefits.

### 5.2. Functional unit

The purpose of the functional unit is to provide a quantitative reference that represents the product or process analysed together with its functionality (i.e., performance). Therefore, a comparative assessment of different products shall always include the fundamental characteristics and performances of the products themselves. The selection of a proper functional unit for concrete materials was already discussed by Marinković et al. (Marinković et al., 2023). The suggested approaches vary according to the goal of the LCA study. For comparative assessments at material scale, the unit volume should be integrated with performance parameters such as the compressive strength. However, the material is meant to be applied in a concrete structure, whose performance requirements are associated to its service life. The upscaling to a real structural element might change the influence of the material properties on the durability, thus the element itself should be adopted as functional unit.

Four of the five considered studies referred the assessment to one cubic meter of concrete (Monkman, 2018; Zhang et al., 2024; Monkman and MacDonald, 2017; Monkman et al., 2023), while the remaining one considered a wall made of 1125 concrete blocks (Monkman and MacDonald, 2016). Together with the unitary volume, each study included different properties for the definition of the functional unit. In (Monkman and MacDonald, 2017) the comparative assessment was based on a reference mix with 27.6 MPa compressive strength at 28 days. Conversely, in (Monkman, 2018) the target strength was not explicitly defined; however, the comparison was made under the assumption that the baseline production and the carbon-mixed concrete had the same performance. Finally, Monkman et al. (Monkman et al., 2023) and Zhang et al. (Zhang et al., 2024) introduced another index, referred to as "Carbon intensity" and "CO<sub>2</sub> index", both defined as the ratio between the carbon dioxide emissions per cubic meter of concrete (kgCO<sub>2</sub>-eq./m<sup>3</sup>) and the compressive strength of the material (MPa) (Damineli et al., 2010). For the purpose of comparing the carbonated concrete with a baseline product, the functional unit shall include the unitary volume and its most relevant property, which for concrete can be assumed to be the compressive strength.

### 5.3. Impact categories

Most of the studies only assessed the carbon footprint associated with the carbon mixing concrete, thus calculating the balance between the

emitted and absorbed CO<sub>2</sub> as well as the savings enabled by the performance enhancement due to the enforced carbonation (Monkman, 2018; Zhang et al., 2024; Monkman and MacDonald, 2017; Monkman and MacDonald, 2016). Contrarywise, Monkman et al. (Monkman et al., 2023) conducted a life cycle assessment with six life cycle impact categories from the ReCiPe model (Huijbregts et al., 2017), namely global warming potential (GWP), ozone depletion (ODP), acidification potential (AP), eutrophication potential (EP), smog (SFP), and non-renewable energy (FFD). As a result, the impact assessment showed that integrating carbon dioxide injection not only helps curbing greenhouse gas emissions but also reduces detrimental environmental effects from a broader perspective. Specifically, the two investigated mixes achieved a reduction of 2.6 %-6.0 % based on the impact category considered (Monkman et al., 2023). The GWP is a relevant indicator for the carbon capture, utilization, and storage (CCUS) technologies, given their focus on carbon dioxide sequestration from the atmosphere. It shall be however calculated with the pertinent unit (i.e., kgCO<sub>2</sub>-equivalent), including all the greenhouse gases, which can be particularly relevant when accounted for in the production of the injection equipment or for the operational costs. Additional impact categories from more comprehensive Life Cycle Impact Assessment (LCIA) models could provide deeper insights into the effects of CO<sub>2</sub> injection on specific impact areas. However, data limitations are likely even more pronounced for categories often excluded in studies, underscoring the need for further data collection to accurately quantify these additional impact indicators. It is also important to consider the inclusion of comprehensive LCIA models based on the study's objectives. While CO<sub>2</sub>-related impacts are central to CCUS, other frequently overlooked indicators may reveal potential downsides to concrete carbonation, such as increased Ozone Depletion Potential (ODP) or Smog Formation Potential (SFP).

### 5.4. Environmental perspectives for concrete carbon mixing deployment

Five studies in the literature assessed the environmental perspective for the scale up of concrete carbon mixing, particularly focusing on the potential of such technology to curb the emissions of concrete production (Thonemann et al., 2022; Ravikumar et al., 2021; Lim et al., 2019; Ravikumar et al., 2021; Silva et al., 2024). Ravikumar et al. (Ravikumar et al., 2021) introduced the climate return on investment (ROI), defined as the ratio between the net climate benefit – accounting for the CO<sub>2</sub> emitted from producing the CCU product and the CO<sub>2</sub> avoided by substituting the conventional process – per functional unit and the CO<sub>2</sub> utilized per functional unit. The use of CO<sub>2</sub> mixing enables a reduction in binder content in the mix, resulting in a 75 % and 70 % likelihood of a positive climate ROI for CO<sub>2</sub> mixing with OPC and OPC with SCMs, respectively. Additionally, the likelihood of having a net CO<sub>2</sub> benefit on 29 concrete carbon mixing cases resulted in 6 out of 8 positive cases for OPC mixes and 19 out of 21 positive cases for OPC + SCMs mixes (Ravikumar et al., 2021), thus confirming the promising development of such technology. The net CO<sub>2</sub> benefit was confirmed by Thonemann et al. (Thonemann et al., 2022) that calculated a global warming potential reduction of 35 kgCO<sub>2</sub>-eq. per kg of CO<sub>2</sub> used for concrete carbon mixing. Finally, Lim et al. (Lim et al., 2019) considered three different mitigation scenarios that included both the CO<sub>2</sub> sequestration through mixing (1.09 %) and the mix design optimization enabled by the injection: a 58.7 %, 37.9 %, and 10.8 % reduction was calculated for optimistic, realistic, and pessimistic scenarios respectively, defined according to the development and availability of supplementary cementitious materials to replace OPC. It is however worth noting that the savings are mostly derived from the reduction in the binder content that the CO<sub>2</sub> injection facilitates, as also confirmed in the case study by Monkman and MacDonald (Monkman and MacDonald, 2017).

## 6. Current economic framework and upscaling challenges

The economic assessment is a crucial aspect for the industrial

deployment of a new technology. The implementation of additional equipment in the concrete production streamline might increase the costs and thus offset the beneficial effects of the carbon dioxide mixing on the performance of the final product. Firstly, the costs associated with the carbon capture and delivery shall be considered. Rajabloo et al. (Rajabloo et al., 2023) provided a summary of the costs related to the CO<sub>2</sub>, specifically the capture costs from cement plants and direct air capture (DAC) technologies, the transportation and storage of the liquefied CO<sub>2</sub> and the truck transportation (Table 10), with reference to the European market.

Four of the reviewed studies reported the market cost of the CO<sub>2</sub>, which is comparable to the data collected by Rajabloo et al. (Rajabloo et al., 2023), as shown in Table 10. Three of the studies assessed the economic viability of using carbon dioxide injection as an accelerator and/or admixture to enhance the performance and sustainability of the final product. Specifically, Monkman and MacDonald (Monkman and MacDonald, 2016) calculated a net sequestration of 1.4 % of the cement emissions in a concrete blocks wall with an additional cost ranging between  $0.85\text{--}1.14 \times 10^{-3}$  € per block (+1% of the current price). On the other hand, Lippiatt et al. (Lippiatt et al., 2019), Monkman et al. (Monkman et al., 2016), and Li et al. (Li et al., 2023) suggested the use of carbon dioxide as set-accelerator, although the latter did not indicate any specific economic calculation. On average, the cost of a common accelerant is approximately 4 \$ / kg, with a demand of 20 kg of accelerant per ton of concrete produced (Lippiatt et al., 2019). The solution proposed by Lippiatt et al. (Lippiatt et al., 2019) requires only 9 kg / ton of CO<sub>2</sub>, with a total cost of 9 \$ / ton (−89 %), while Monkman et al. (Monkman et al., 2016) reported a total cost of 0.48–2.85 \$ for a truckload of concrete (8 m<sup>3</sup>), significantly abating the costs of a traditional calcium nitrate accelerant (12.36–24.72 \$, −91 % on average). Athar Kazmi et al. (Athar Kazmi and L. Vara Prasad Meesaraganda, P. Suresh Chandra Babu, , 2023) proposed a simplified cost analysis introducing the economy index to compare the performance of the four investigated concrete mixes – the reference mix and three different CO<sub>2</sub>-injected mixes with variable dosage. The economy index (EI) was defined as the ratio between the compressive strength evaluated at 28 days, and the total cost of the raw materials, including the CO<sub>2</sub> for the carbonated mixes. As a result, the economy index increased for all the three carbonated concretes from the reference value of 0.80 up to 0.83, indicating the efficiency of the sequestration process from an economical perspective, with the middle-dosage mix being the most efficient (0.48 kg<sub>CO2</sub> / m<sup>3</sup>, EI = 0.83).

Albeit having specific data at material scale, the industrial upscaling requires additional data on the implementation of injection systems in a production streamline, which is currently lacking. Therefore, only few projections provide a broader perspective on the use of concrete carbon mixing as an economically viable carbon sequestration technology. Lim

et al. (Lim et al., 2019) proposed a comprehensive overview of the scalability of concrete carbon mixing as compared to concrete carbon curing and carbonation of recycled aggregates. From a cost perspective, the savings enabled by the reduced raw material consumption for carbonated concrete can offset the additional costs of CO<sub>2</sub> supply (34–69 \$ / ton<sub>CO2</sub>) and CO<sub>2</sub> transportation and processing (10–2000 \$ / ton<sub>CO2</sub>) if a OPC reduction of 16 % is achieved. In addition, Lim et al. (Lim et al., 2019) suggested that the further promotion of industrial deployment of these CCU technologies is driven by the introduction of financial policies related to CO<sub>2</sub> sequestration, such as the Carbon Border Adjustment Mechanism (CBAM) in the European Union and the 45Q tax credit in the United States.

The promising economic perspective is however substantiated only by a limited number of primary data, while future deployment potential mainly consists of projections based on the industrial implementation of the carbon mixing technologies. It is hence necessary to address the current challenges for the upscaling of laboratory studies. Firstly, reaction kinetics and fluid mechanics are strongly affected by the geometry and size of the environment where the carbonation happens. For instance, high pressures and properly sealed systems are hard to replicate at industrial scale, particularly considering the production volumes of concrete batching plants. Moreover, the different reaction kinetics might impair the efficiency of the carbonation reactions. The injection of gaseous CO<sub>2</sub> would necessarily be implemented from specific spots on the surface of the mixing bowl, making it hard to properly interact with the inner particles if the cementitious mass is not properly circulated in the system. Additionally, mixing times might strongly vary depending on the volume treated with the injection system, and the experimental data might not be applicable.

## 7. Conclusions

This study reviewed the state of research on concrete carbon mixing technology. A systematic literature review identified three retrofit routes for carbon dioxide fixation in concrete production: carbonation of the mixing water before it enters the main mixer, injection after the binder is added, or injection with all ingredients already in the mixer. Key parameters for each method were identified, and the effects of early carbonation on concrete properties were compared across the existing studies. The study also addressed methodological aspects of environmental assessment, focusing on life cycle assessment (LCA), and discussed the economic scalability of concrete carbon mixing.

In conclusion, the following key points summarize the main remarks of the present review:

- The use of carbonated water showed that it could enhance the compressive strength of concrete (up to + 20 %) while complying with the existing standards on the minimum pH of mixing water. The upscaling of highly pressurized systems might however be particularly challenging from an industrial perspective due to the implementation of sealed components.
- The injection of CO<sub>2</sub> in the cement slurry, if limited to short injection times, can increase the compressive strength of the carbonated paste (up to + 18 %), while longer carbonation times might be detrimental for the microstructure development. Nonetheless, when transitioning from laboratory-scale technology to industrial plants, the upscaling might affect the reactions during injection, thus requiring further investigation at larger scales.
- Direct carbon dioxide injection in concrete demonstrated the accelerating effects of carbonation, significantly reducing both the slump and final setting time by an average of 22.9 % and 20.2 %, respectively. Nevertheless, the mechanical performance, mainly assessed through compression tests, can be enhanced with small CO<sub>2</sub> dosages (up to 0.2 % CO<sub>2</sub> dosage by weight of binder); at higher dosages, the effect of carbon dioxide injection might hinder the strength development of the concrete matrix.

**Table 10**

Carbon capture costs associated with concrete carbon mixing (adapted from (Rajabloo et al., 2023) and CO<sub>2</sub> market costs considered by the reviewed studies.

| Process                                                                       | Cost                                                |
|-------------------------------------------------------------------------------|-----------------------------------------------------|
| Cement plant                                                                  | 92–172 € / ton <sub>CO2</sub> (Uibu et al., 2021)   |
| Direct air capture (DAC)                                                      | 200–1000 € / ton <sub>CO2</sub> (Van Dael, 2018)    |
| CO <sub>2</sub> transportation and storage                                    | 10 € / ton <sub>CO2</sub> (Roussanaly et al., 2021) |
| Truck transportation                                                          | 0.22 € / (ton <sub>CO2</sub> × km) (Van Dael, 2018) |
| Study                                                                         | Market cost considered                              |
| (Lippiatt et al., 2019)                                                       | 1000 \$ / ton <sub>CO2</sub>                        |
| (Athar Kazmi and L. Vara Prasad Meesaraganda, P. Suresh Chandra Babu, , 2023) | 370 \$ / ton <sub>CO2</sub>                         |
| (Monkman and MacDonald, 2016)                                                 | 330–440 \$ / ton <sub>CO2</sub>                     |
| (Monkman et al., 2016)                                                        | 385 \$ / ton <sub>CO2</sub>                         |

- Carbonated concrete demonstrated the benefits of CO<sub>2</sub> injection on durability, maintaining pH well above the corrosion threshold, while reducing chloride penetration and permeability by an average of 8.3 % and 8.6 %, respectively.
- A lack of methodological standards due to the novelty of the technology limits the understanding of the environmental performance of concrete carbon mixing; however, as observed in most instances, the benefits of the process appear to result from the binder reduction enabled by CO<sub>2</sub> injection, rather than from CO<sub>2</sub> sequestration.
- Preliminary studies suggest that carbon dioxide can be used in concrete as a cost-effective, sustainable set-accelerator (up to 90 % cheaper), but further evaluation is needed to assess the overall process, including capture costs and the evolving CO<sub>2</sub> market.

Future work should focus on further investigation on the correlations between the process parameters and the properties of carbonated concrete. Specifically, the performance assessment should delve into multiple aspects that are crucial for concrete use in structural application, including fresh state, mechanical (i.e., compressive and tensile strength, fracture toughness), and durability properties.

At present, the performance enhancement is only limited to small quantities of CO<sub>2</sub> sequestration. Therefore, systematic LCA studies with broader LCIA models should determine to what extent concrete carbonation in the mixing phase positively affects the emissions of concrete industry. Moreover, the laboratory scale experiments should be the driver for further development towards bigger scale applications, which might pose a technological challenge due to the upscaling of the operational equipment, with specific regards towards reaction kinetics of higher production volumes. To conclude, a comprehensive understanding of the input–output relationship for carbon dioxide injection during the mixing phase could significantly enhance the adoption of this technology in ready-mix concrete plants and precast concrete factories. Additionally, if more compact batching systems are developed, it could facilitate the use of this technology at construction site batching plants.

#### CRedit authorship contribution statement

**Marco Davolio:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Giovanni Muciaccia:** Writing – review & editing, Supervision, Conceptualization. **Liberato Ferrara:** Writing – review & editing, Supervision, Conceptualization.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgements

The work reported in this paper is part of the stakeholder outreach activity performed in the framework of the project MUSA – Multilayered Urban Sustainability Action – project, funded by the European Union – NextGenerationEU, under the National Recovery and Resilience Plan (NRRP) Mission 4 Component 2 Investment Line 1.5: Strengthening of research structures and creation of R&D “innovation ecosystems”, set up of “territorial leaders in R&D”. The discussions and exchange of ideas with Federico Furlani and Stefano Gelain (SIMEM S.p.a.) have been especially appreciated.

#### Data availability

Data will be made available on request.

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