

Review

Strategies for OPC Paste Carbonation: Relationship between Microstructure, Performance and Net CO₂ Balance

André Silva *, Rita Nogueira  and José Alexandre Bogas 

Civil Engineering Research and Innovation for Sustainability, Department of Civil Engineering, Architecture and Environment, Instituto Superior Técnico, Universidade de Lisboa, 1049-001 Lisboa, Portugal; rita.nogueira@tecnico.ulisboa.pt (R.N.)

* Correspondence: andre.f.rosario.marcelino.silva@tecnico.ulisboa.pt

Abstract: Carbon capture storage and utilization is the main technology for reducing CO₂ emissions, accounting for 56% of the overall reduction required to achieve the carbon neutrality of concrete by 2050. Different strategies have been explored in cement-based materials towards this end, namely, in concrete. However, the impact on carbonated concrete differs depending on the moment at which cementitious material comes into contact with CO₂, either in terms of CO₂ uptake or in terms of its lifetime performance. This paper presents three leading strategies that rely on the direct carbonation of a cementitious binder to reduce the carbon footprint. For each strategy, the effect of the carbonation process on the kinetics and microstructure of cementitious paste, the estimation of its carbon capture capability and the application feasibility are discussed. Accelerated carbonation curing is one approach widely studied by academics. However, despite some CO₂ capture effectiveness, its industrial processing is still a long way off. A second strategy consists of incorporating CO₂ during the mixing process, which has been shown to speed up the hardening reactions of cement. However, this effect is of short term and may negatively affect its long-term performance. Finally, the carbonation of hydrated cement waste is shown to be a very promising strategy that enables the recycling of hydrated cement waste as a supplementary cementitious material which also has a potentially high CO₂ uptake. The integrated analysis of the three strategies highlights a wide variability in the reduction of CO₂ emissions from 1% to 37% in relation to current emissions, where the best result was achieved using carbonated waste (third strategy) in the production of a concrete subjected to carbonation curing (first strategy).

Keywords: carbon capture utilization and storage; precast concrete industry; carbonation curing; CO₂ uptake; cement paste waste



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1. Introduction

Presently, concrete is preponderant in the development of modern societies, being the most used construction material with the highest annual volume consumption after water, justified by its excellent cost–performance relation [1–3]. However, the production of concrete accounts for approximately 8% of global carbon dioxide (CO₂) emissions. Moreover, concrete consumption is expected to increase by 2050 to fulfil the requirements of modern societies. Accordingly, the demand for ordinary Portland cement (OPC) is estimated to increase by 20% in the same period, further worsening CO₂ emissions due to its large carbon footprint [2,4].

Each operation of concrete production, from extraction of raw materials to its application, contributes to the CO₂ emission issue. However, the most impactful stage reoccurs during the cement manufacturing process, which is responsible for around 76% of total CO₂ emissions, by mass [2,5–7], Figure 1. Naturally, there is also the important effect of concrete carbonation during its life cycle and after decommissioning. However, this process during these two periods only represents, on average, about 16% and 1.5% of concrete-related

CO₂ emissions, respectively. However, these values only correspond to around 38% of the concrete carbonation potential (assuming an average content of CaO in cement of 60%), exposing the possibility of further carbonation [5,8]. These percentages can be translated into the partial values of CO₂ emissions/absorptions indicated in Figure 1, assuming a total emission of almost 300 kg of CO₂ per cubic meter of concrete [2,9,10]. Therefore, achieving the carbon neutrality of concrete still remains an ambitious desire.

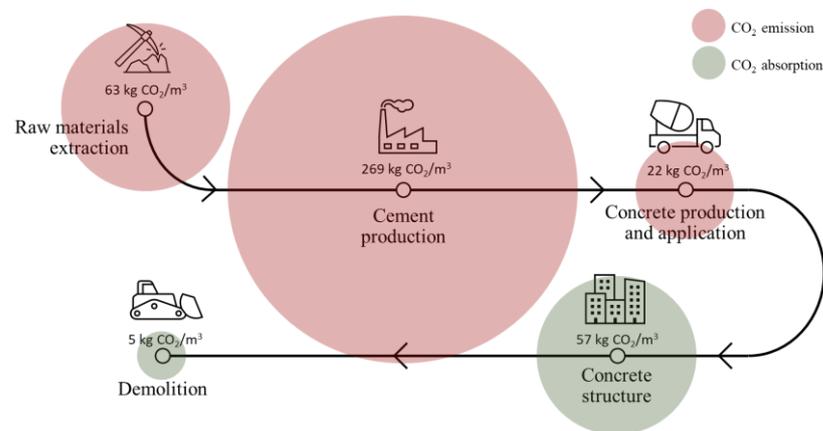


Figure 1. Generic CO₂ cycle of concrete (kg/m³) [2,9,10].

In 2009, the International Energy Agency (IEA) published the Technology Roadmap (Low-Carbon Transition in the Cement Industry), later updated in 2018, which aimed at defining strategies for reducing the CO₂ emissions associated with the cement industry, and consequently with the concrete industry [1,3]. The IEA roadmap analysed a diverse set of carbon mitigation strategies and identified four main levers with higher impacts on carbon emissions: thermal and electrical efficiency; alternative fuels; clinker substitution; and carbon capture utilization and storage (CCUS) [1,3]. Additionally, to achieve the ambitious CO₂ emission reduction target of about 80% by 2050 from 2009 levels, these strategies must be implemented together; however, CCUS technology remains insufficiently explored, especially considering that it accounts for 56% of the planned CO₂ reduction.

Similarly, in 2018, Cembureau complemented a previous roadmap from 2013, aiming at “Reaching climate neutrality along the cement and concrete value chain by 2050” [11], a goal shared with the aforementioned IEA roadmap. Despite the common purpose, Cembureau’s roadmap focused on a broader sector, including the concrete industry in the overall path to carbon neutrality. This roadmap targets the “5C’s”: clinker, encompassing low-carbon clinker, thermal efficiency and biomass fuels; cement, encompassing clinker substitution, renewable energy and electrical efficiency; concrete, refining concrete mix; construction carbonation, engaging the CO₂ capture of concrete in use; and finally, carbon capture utilization and storage, which comprises carbon sink technologies. In this roadmap, Cembureau also considers CCUS technologies as the main strategy for CO₂ reduction along the cement value chain, responsible for about 42% of CO₂ emissions per ton of cement. Furthermore, in 2020, the Global Cement and Concrete Association (GCCA) created an exhaustive roadmap connecting the previous ones, estimating that CCUS technologies will contribute to around 36% of total CO₂ emission savings by 2050 [12].

CCUS technology intends to capture CO₂ followed by the utilization of the obtained carbonated product, or simply to store carbon inhibiting its emission to the atmosphere. This strategy was only recently considered potentially competitive after advances implemented on the governmental level, namely, carbon tax or emission trading systems. Throughout different countries, academia and industry are developing different strategies for the application of CCUS technologies throughout the value chain of both concrete and cement, including CO₂ recovery from flue gas, converting CO₂ into a by-product provided by many industries [13]. Thus, the availability of CO₂ at competitive prices is expected

to increase in the near future, thus promoting the competitiveness of CCUS technologies. Hence, the prospect of CO₂ availability prompts the exploration of new solutions for the efficient reuse of CO₂ [14–17].

The strategy of adopting CCUS technologies in the value chain of concrete has motivated the publication of a wide collection of papers and reports, focused on the revision of different, yet specific, CCUS technologies. Some studies focused on the introduction of CCUS technologies in the cement production stage, summarizing strategies for the separation of CO₂ from flue gas and strategies for clinker substitution [18,19]. Other studies focused on the introduction of CCUS technologies in the concrete production stage, analysing different prospects for the carbonation and utilization of concrete and industrial waste within the concrete composition [20–27].

In contrast, this paper reviews three strategies for the application of CCUS technologies that directly affect the cementitious binder, i.e., direct carbonation of the cementitious binder. Since, in each strategy, the exposure to CO₂ occurs in different stages of concrete production, this approach enables a comparative analysis of the effects of carbonation on the microstructure of the cementitious paste, some of them eventually deleterious to the concrete performance. This analysis is considered the biggest value addition of this paper as it sheds light on a hitherto unexplored topic. Moreover, the carbon capture capability and the application feasibility of each strategy are also addressed, enabling a broader view of the carbonation potential of each strategy.

The three strategies for CCUS technologies in concrete production reviewed in this paper are illustrated in Figure 2. The first strategy consists of the carbonation curing of precast concrete, a process that has already proved its CO₂ capture effectiveness, but with its industrial viability yet to be demonstrated. The second strategy is the carbonation during the mixing stage, which has been proven to speed up the hydration reactions of cement at an early stage, while introducing the risk of a negative impact on its long-term performance. The third strategy discussed here is the carbonation of hydrated cement waste from demolished concrete, an approach that presents the highest carbonation potential among the three and promotes both the circular economy and clinker substitution by enabling the recycling of end-of-life concrete as a filler addition. Finally, an overview of these three strategies is presented, comparing their main distinctive characteristics, industrial viability and net CO₂ balance. For each strategy, the pros and cons are discussed, the main findings reported in the literature are presented and the feasibility of mitigating carbon emissions is evaluated. Thus, this paper intends to reveal the state of the art of concrete carbon capture strategies, aiming at reducing the carbon footprint of concrete production.

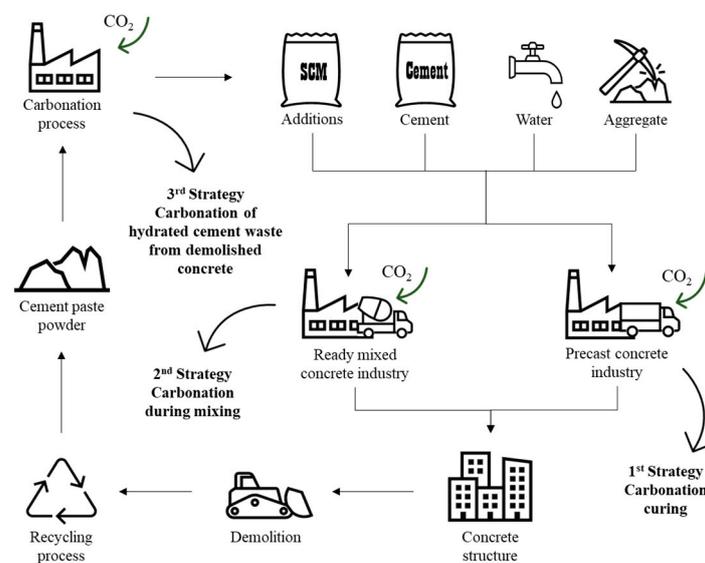


Figure 2. CCUS strategies present in the concrete life cycle.

2. Overview of Hydration and Carbonation of Cement-Based Materials

Cement is a critical concrete component, affecting its physical, mechanical and durability properties, even though it only corresponds to about 13–18% of ordinary concrete mass [6]. Besides concrete, this hydraulic binder has a wide diversity of applications in the construction sector, namely, in mortar and other cement-based materials.

The manufacturing process of cement comprises a diverse set of complex steps, from extraction of raw materials to the final Portland cement product. The raw materials are typically limestone, sand and clay (or marl) as sources of CaO, SiO₂ and Al₂O₃, the main cement components. The ground raw materials are blended in predefined proportions and heated at about 1450 °C in the pyro processing stage. This stage is responsible for around 85% of the total carbon emissions from the cement industry (≈60% from limestone calcination and ≈40% from fuel combustion) [2,5,8,28]. The obtained product (clinker) is mainly composed of tricalcium silicate, C₃S (45–65%); dicalcium silicate, C₂S (10–30%); tricalcium aluminate, C₃A (5–12%) and tetracalcium aluminoferrite, C₄AF (6–12%) [29]. The final cement product is obtained from the milling of clinker until a particle size reduction to about 10 μm is reached, on average, and the addition of gypsum [30–33].

2.1. Hydration of Cement-Based Materials

The binding effect of the cementitious matrix results from the hydration reaction of the cement anhydrous compounds, yielding a densely cross-linked phase of calcium silicate hydrates (C-S-H) and other minor hydration products, mainly calcium hydroxide (CH) and calcium aluminate hydrates [34]. C₃S, the major cement compound, quickly dissolves into the liquid phase once in contact with water, forming C-S-H and CH, following Equation (1) [34]. The hydration reaction of C₂S is similar, but with a much slower rate [34]. Concerning the aluminate phase (C₃A and C₄AF), the hydration mechanism is similar, differing in the reaction rates throughout the full extent of the hydration process. Upon contact with water, C₃A immediately dissolves into the liquid phase and quickly hydrates. In the presence of a source of calcium sulfate (G), this reaction produces ettringite (AFt), a needle-like phase. After G is depleted by the aluminate reaction, AFt reacts with additional amounts of C₃A to form monosulfate aluminate (AFm) [35–37].

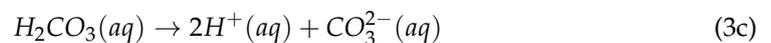
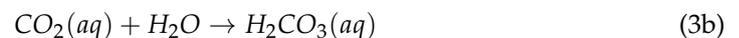


Cement hydration kinetics are commonly divided into four periods: the pre-induction, induction, acceleration and deceleration periods. The pre-induction period is primarily characterised by a surface dissolution of the anhydrous cement compounds in reactive sites, originating a short-lived hydration reaction with a simultaneous heat release that quickly decreases within a few minutes [34,37–39]. The induction period begins with a drastic reduction of the hydration rate. The reasons for this reduction are not fully understood, and several theories have been proposed. The main theories that have earned more recognition are the protective membrane theory and the dissolution theory [37]. The first theory advocates that the formation of a protective layer of hydration products on the surface of anhydrous cement particles hampers the anhydrous phase dissolution, thus reducing the hydration rate [34,40]. According to this theory, the induction period ends once the hydrated layer is destroyed or rendered more permeable as a result of ageing, phase transformation or even osmotic pressure. The dissolution theory states that the reduction in the hydration rate is ruled by the increase in the concentration of calcium and hydroxide ions in the liquid phase [36,40]. This phenomenon generates a reduction of the undersaturation level of anhydrous phases, which lowers the dissolution rate and consequently the hydration rate [39]. The hydration products, namely, C-S-H and CH, eventually begin to nucleate and grow [41]. The concentration of ions in the liquid phase starts to reduce, reverting the abovementioned reduction trend in the undersaturation level of anhydrous phases. Consequently, the dissolution of anhydrous compounds accelerates, causing an increase in the hydration rate and ending the induction period after around

3–4 h from contact with water [36,40]. In the acceleration period, which lasts for about 4–6 h, both C-S-H and CH grow rapidly. Finally, in the deceleration period, there is a reduction in the hydration rate that is attributed to the lack of porous space for the growth of new hydration products. Even though the hydration reaction continues for many days, the hydration degree is usually evaluated after 28 days, at the moment when the hydration reaction has reached about 80% [38–40].

2.2. Carbonation of Cement-Based Materials

As previously mentioned, concrete undergoes a natural carbonation process during its life cycle, partly offsetting its high carbon footprint. This process begins at the exposed surface, progressing inwards in a slow and diffusion-controlled process [42]. The carbonation reaction does not cause significant deterioration of Portland cement concrete; in fact, the literature demonstrates that there is usually a porosity reduction caused by the deposition of carbonate compounds [43,44]. However, there are indeed durability issues related to the negative impact of carbonation in reinforced concrete. Prior to the carbonation reaction, the steel rebars are protected against corrosion by a passive layer of oxides, creating the high pH (12.6 to 13.5) of the cement matrix pore solution [43,44]. This high alkalinity is mainly maintained by the CH which provides hydroxide and calcium ions into the liquid phase following Equation (2). During carbonation, CO₂ dissolves into the liquid phase (Equation (3a)) to form carbonic acid, H₂CO₃ (H₂C^o) (Equation (3b)), also originating carbonate and bicarbonate ions (Equation (3c)). Afterwards, the bicarbonate ions react with calcium ions, also present in the liquid phase, creating calcium carbonate (CC^o) according to Equation (4), and lowering the pH close to the steel bars [16,45].



The kinetics of natural concrete carbonation are mainly controlled by the ability of CO₂ to diffuse through the cement paste pore system. The amount of water inside the pores is a crucial factor in the carbonation rate. The diffusion of CO₂ in water is four orders of magnitude slower than in air. Conversely, in the absence of water, CO₂ remains gaseous and unable to carbonate, following Equation (3). Thus, the relative humidity conditions required to maximize the carbonation rate fall within the range of 50–70%. Furthermore, the inward movement of the progressive carbonation is increasingly hampered by the previously carbonated layer, which became denser due to deposition of the carbonate precipitates. The carbonation reaction also releases water, as expressed by Equation (4), which might either aid or hamper the carbonation reaction [31,46].

The carbonation potential of concrete relies on the availability of reactive calcium in the different compounds of the cement paste. The complete carbonation of this calcium content would amount to 165 kg of CO₂ per m³ of a concrete with a dosage of 350 kg of cement per m³, which corresponds to 61% of the total CO₂ produced in the cement manufacture. Afterwards, the natural carbonation of concrete that occurs during all the different stages of service life, from application to demolition and concrete waste treatment, might reduce this amount to an estimate of 103 kg of CO₂ per m³ of concrete (38% of the CO₂ released). Hence, the potential for further carbonation technologies is a reality which has motivated recent studies concerned with high carbon emissions from the cement industry.

Thus, industrial processes have been increasingly studied and developed with the aim of achieving full carbonation of cementitious materials and consequently maximizing the CO₂ uptake. Nevertheless, conditions for a feasible industrial carbonation process are far from being established. Although it is generally accepted that the concept of

forced carbonation (or accelerated carbonation) is preferable to natural carbonation, the uncertainty associated with the reaction products and with the kinetics and mechanisms of the carbonation reaction makes it difficult to define a generic carbonation process. Note that both anhydrous and hydrated cement compounds are carbon-reactive with, however, different carbonation potential and ideal conditions for the reaction. C_3S and C_2S are the main carbonatable reactants in cement, forming CC of different crystallinity degrees and amorphous silica in the presence of water. The carbonation reaction of C-S-H and CH also produces CC and amorphous silica for a high-enough carbonation level. Hence, this process causes the decalcification of C-S-H and the polymerization of its structure. Moreover, besides C-S-H and CH, AFt and AFm also react with CO_2 . While the reaction of AFt with CO_2 produces G, CC and alumina gel, the reaction of CO_2 with AFm leads to the formation of monocarbonate aluminate (Mc) and hem carbonate aluminate (Hc) [16,35,47,48].

3. Carbonation Curing of Cementitious Materials

In this strategy, CO_2 is applied to the cementitious material during the curing process, after mixing and casting. The cast specimens harden during this stage, and carbonation has the potential to accelerate the strength development. The carbonation curing strategy is essentially limited to the precast industry. Being intrinsically a process that occurs over time, the curing stage is of crucial importance to the competitiveness of this industry [49]. Thus, its implementation promotes product turn-over, leading to improvements in process efficiency and productivity [17,50].

Currently, some precast companies use steam during the curing stage to obtain an environment with a high temperature (T) of up to 70 °C, and a high relative humidity (RH), above 95%, in a very energy-intensive process. Even though CO_2 was proposed in the 1970s as an alternative source for the curing stage, the high production cost and some reported durability problems hampered its use on a larger scale [50]. However, the recent focus on the mitigation of greenhouse gas emissions has restored interest in this strategy. Moreover, the expected increase in CO_2 availability and progress in carbon capture technologies also contributed to the reestablishment of this line of investigation. In fact, the utilization of CO_2 as a feedstock for cementitious material curing can potentially provide not only a positive environmental impact but also economic benefits with similar or even lower costs than those of steam [51]. Nevertheless, a generic feasible curing process using CO_2 has not yet been defined, since, as briefly mentioned, an effective carbonation process is supported by specific conditions (curing atmosphere and material characteristics) that are not yet fully established [17,52,53].

Precast concrete comprises dry-mix and wet-mix compositions. Dry-mix consists of the common small non-structural products, e.g., masonry units or paving stones, and is characterized by a very stiff fresh mix, with a low w/b ratio and a moderate to low cement content. With the casting and mechanical compaction by an external force conducted simultaneously and avoiding formwork and demoulding, the manufacturing process of dry-mix products is inexpensive. However, the final product usually presents a poor appearance (irregular and sometimes cavernous), limiting its range of application. The wet-mix materials are formulated with a higher w/b (usually above ≈ 0.3) and/or plasticizer, creating a more fluid material. These materials require formwork and can only be demoulded after a few hours. This process leads to products aimed at decorative purposes and structural elements [54]. The following sections discuss the carbonation process, main reactions and phase development, material performance and CO_2 uptake of the carbonation curing strategy.

3.1. Carbonation Curing Process

A generic carbonation process aimed at the curing stage of cementitious materials is generally composed of three stages: pre-conditioning, carbonation and post-conditioning [52,55] (Figure 3). The purpose of the pre-conditioning stage is to reduce the pore water content in the porous structure to improve CO_2 diffusion through the cementitious matrix during

the following carbonation stage. This can be avoided in dry-mix concrete, since the water content inside its pores is sufficiently low. Conversely, in wet-mixes, the pre-conditioning stage is crucial and is carried out under relatively dry conditions, usually 20–25 °C and 40–60% RH. The conditions and duration of this stage will affect the final performance of the mixes [56]. Note that the absence of a pre-conditioning stage in dry-mix concrete results in the beginning of the carbonation stage upon the induction period of hydration. This condition is known to have an accelerating effect on the hydration reactions, contributing to a higher early-age mechanical strength, which in turn promotes the product turn-over [57–59]. On the contrary, the application of a pre-conditioning stage a few hours after production (after demoulding), as in wet-mix concrete, moves this carbonation process to the acceleration or even the deceleration period of hydration. As a consequence, the contribution of carbonation to the earliest mechanical properties is hampered. Additionally, the drying conditions of the pre-conditioning stage cannot be too severe, ensuring a balance between hydration and carbonation reactions. A strong drying phase will favour subsequent carbonation in the carbonation stage but will penalize hydration, thus compromising the future performance of the material [17,52].

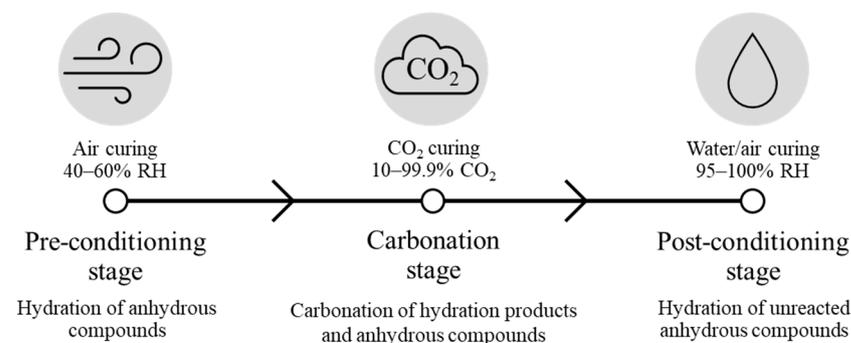


Figure 3. Stages of carbonation curing [52].

Afterwards, in the carbonation stage, the cementitious materials are carbonated in a process that can be performed in either an enclosed or flow-through system. These processes differ in energy consumption and carbonation efficiency, but both adopt environmental conditions of 60–70% RH and 20–40 °C for effective carbonation. Studies have shown that the carbonation degree is maximized at around 50% RH, with poorer performances for both higher and lower RH values [60]. However, this decrease in the carbonation degree is relatively low in the range of 50–65% RH, which is the reason why the ambient environment is usually applied [21,48,61]. The temperature factor also presents a limited impact on the carbonation degree for the 15–80 °C range, since higher temperatures accelerate water evaporation and reduce CO₂ solubility in the liquid phase. As a consequence, the carbonation is usually conducted at room temperature [52,62,63]. Concerning the carbonating atmosphere, two parameters are usually considered: CO₂ concentration and partial pressure. Generally, studies use pure CO₂ gas with partial pressures of 1–5 atm and CO₂ concentration ranging from 10 to 99.9%, although flue gas can be used instead for a more economical and eco-friendly procedure [48,64]. These parameters affect the ingress, diffusion and dissolution of CO₂ in the liquid phase of the cementitious matrix, being both beneficial for CO₂ mobility and alkali metal ion leachability. However, an intensive carbonation, i.e., high CO₂ concentration and partial pressure, can result in the formation of a passive \overline{CC} layer on the surface of cementitious particles blocking the contact between CO₂ and uncarbonated material and simultaneously prompting the development of microcracking due to the large amount of heat generation [61,65]. Similarly, the duration of the carbonation stage also leads to a positive influence on the carbonation degree. Yet, the carbonation rate reduces over time, with studies pointing to a 10 h threshold above which the increase in CO₂ uptake becomes negligible [51,61].

In the final stage, post-conditioning, the cementitious materials are cured in moist conditions, usually with $RH \geq 95\%$ or even in water immersion. The objective is to promote the hydration of the remaining unreacted cement phases, simultaneously mitigating the reduction in the pH of the pore solution [17,52,54].

Besides the specific set of carbonation conditions, for efficient carbonation curing to be accomplished, the intrinsic physical characteristics of the material (e.g., microstructure) must also be discussed and defined. On this matter, a connected porosity in the cement paste matrix increases permeability, which consequently facilitates the CO_2 diffusion and the carbonation of the cement reactive compounds. While this condition is easily achieved in the porous microstructure of dry-mixes, for wet-mixes this purpose is usually achieved by raising the w/b ratio. However, this increase is limited to a w/b ratio of 0.55, since above this limit the cement reduction in the concrete volume hinders the availability of carbon-reactive compounds, compromising the viability of the strategy [66]. Note that the CO_2 diffusion mechanism is continuously hindered by the precipitation of carbonates in the porous space of external layers.

Similarly, the pore moisture content must be controlled to enable both CO_2 diffusion and CO_2 dissolution, an issue addressed during the pre-conditioning stage. Depending on the w/b (from 0.34 to 0.5), maximum carbon absorption was reported for water removal between 4.5 and 30% of the initial mixing water [52,67]. While wet-mixes with higher w/b require a lower amount of water removal to create sufficient space for CO_2 diffusion, in dry-mixes, the opposite occurs. The higher w/b originates mixes with coarser pores which provide the needed air space availability for the diffusion of CO_2 even if partially filled with water. Conversely, the dry-mixes need more extensive water removal to offset the smaller size of the pores [52,54].

Moreover, the particle size of cement grains must be considered, since the higher specific surface of cement particles promotes the exposition to carbonate pore water of the carbon-reactive compounds, following Equations (3) and (4) [52,68]. Table 1 summarizes the results of compressive strength improvement and CO_2 uptake reported in recent works.

Table 1. Compressive strength and CO_2 uptake of cement-based mixtures after carbonation curing.

Mixture	Process	Compressive Strength Variation			CO_2 Uptake (%wt of Clinker)	Refs.
Wet-mix concrete (w/b = 0.45)	PC ¹ : 20 h/26 °C/50% CS ² : 6 h/99%/0.69 atm after vacuum	+26% at 3 days +5% at 28 days			14.1%	[69]
Wet-mix concrete (w/b = 0.4)	PC ¹ : 0 h/25 °C/50% PC ¹ : 4 h/25 °C/50% PC ¹ : 18 h/25 °C/50% CS ² : 4 h/100%/1 atm	1 day	7 days	28 days	7.5% 21.3% 24.2%	[56]
		−17%	44%	41%		
		−46%	−13%	−8%		
		−13%	15%	−1%		
Dry-mix paste (w/b = 0.15)	CS ² : 2 h/1%/1 atm CS ² : 2 h/3%/1 atm CS ² : 2 h/10%/1 atm CS ² : 2 h/20%/1 atm	3 days			9.6% 13.2% 16.6% 19.5%	[70]
		+1.2%				
		+3.8%				
		+3.7%				
Dry-mix mortar (w/b = 0.15)	PC ¹ : 0 h PC ¹ : 5 h PC ¹ : 11 h PC ¹ : 23 h PC ¹ : 71 h CS ² : 1 h/99%/1 atm	28 days			9.8% 9.0% 7.9% 6.8% 6.1%	[57]
		−3.2%				
		−3.2%				
		+13.5%				
		+12.8%				
		+37.8%				

Table 1. Cont.

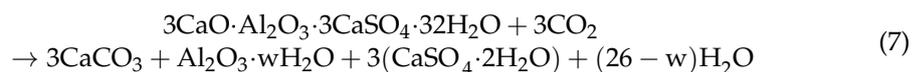
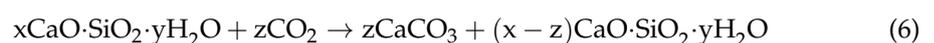
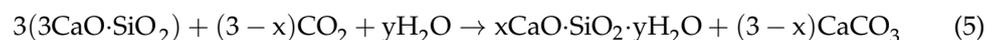
Mixture	Process	Compressive Strength Variation			CO ₂ Uptake (%wt of Clinker)	Refs.
Dry-mix mortar (w/b = 0.3)	PC ¹ : 2 h/60%/20 °C	3 days	7 days	28 days		[71]
	CS ² : 1 h/100%/1 atm	+9.0%	+4.5%	+0.0%	19%	
	CS ² : 5 h/100%/1 atm	+10.5%	+4.5%	−10.7%	21%	
	CS ² : 12 h/100%/1 atm	+15.0%	+9.1%	−10.7%	23%	
Dry-mix paste (w/b = 0.18)	PC ¹ : 2 h		+212.5%		7.9%	[72]
	CS ² : 2 h/20%/1.5 atm		+1.4%		17.6%	
	CS ² : 672 h/20%/1.5 atm	at the end of CS				
Dry-mix paste (w/b = 0.15)	CS ² : 2 h/99.5%/1 atm	3 days +17%	28 days +8%		7.3%	[73]

¹ Pre-conditioning stage: duration/temperature/HR; ² Carbonation stage: duration/CO₂ concentration/relative pressure.

3.2. Reactions and Phase Development

As aforementioned, the reactions and newly arising compounds depend on the mix design and the consequent conditions and duration of the three main stages of the curing process. The hydration reactions begin in the casted sample and last for a relatively short duration, up to 24 h; hence, mainly C₃S and C₃A react during this stage, originating C-S-H, CH and AFt. Note that for a pre-conditioning stage duration not higher than 2 h, the carbonation stage will affect the hydration kinetics during the low reactivity induction period, in which the dissolution of anhydrous cementitious material is slow. If carbonation is delayed for 4–10 h, it will coincide with the period of highest reactivity, the acceleration period, in which the dissolution of anhydrous material is higher [36,40]. These very different conditions will surely have an impact on the hydration reactions and, consequently, on the final performance, as shown in Table 1 [56–59].

Regardless of the pre-conditioning stage duration, the anhydrous cement compounds, being present in higher relative amounts in the carbonation stage, are the main CO₂ reactants, namely, C₃S and C₂S. Note that C₃A is also present at this stage; however, this compound records little reactivity to CO₂. In fact, Wang et al. [74] reported that the CO₂ absorbed by C₃A and C₄AF is small and can be ignored when compared with C₃S and C₂S. As previously mentioned, the carbonation products are silica gel and C \bar{C} with different crystallinity degrees (Equation (5)). Moreover, the C \bar{C} polymorph products obtained during the carbonation stage are influenced by both the intensity of carbonation conditions and chemical composition of the cementitious materials. All the existing C \bar{C} polymorphs have been shown to emerge within the carbonation stage, namely, calcite, vaterite, aragonite and amorphous C \bar{C} . However, calcite, the most thermodynamically stable polymorph, arises as the most common C \bar{C} product [17,47,61]. Additionally, the hydration products can also react with CO₂, namely, CH (Equation (2)), C-S-H (Equation (6)) and AFt (Equation (7)).



The C \bar{C} precipitates from carbonation can act as nucleation sites for further hydration reaction products, thus also contributing to the acceleration of hydration reactions [75,76]. Hence, even though the carbonation stage mainly includes carbonation reactions, the hydration reaction is also promoted and accelerated, with both phenomena occurring simultaneously. By the end of the carbonation stage, the casted sample is mainly composed of C \bar{C} , decalcified C-S-H, silica gel and unreacted cement compounds. Equations (5) and (6) represent the overall reactions between CO₂ and C₃S and C-S-H, respectively. However,

these are dissolution–precipitation reactions with some intermediate coupled processes, namely, the CO_2 dissolution illustrated in Equation (3). Note that $x = z$, in Equation (6), represents extended carbonation where all C-S-H is decalcified to yield silica gel [17,52].

In the post-conditioning stage, further water curing re-establishes the hydration reactions of the remaining unreacted cement compounds, originating additional amounts of C-S-H as well as AFt and AFm phases [77]. The less stable forms of $\text{C}\bar{\text{C}}$ still present, including vaterite and aragonite, eventually convert into calcite. Moreover, the presence of $\text{C}\bar{\text{C}}$ during further hydration also releases carbonate ions to the liquid phase, leading to the formation of Mc and Hc by the eventual replacement of sulphate ions in AFm [48,52].

3.3. Carbonation Impact on Microstructure and Performance

The introduction of a carbonation stage in the conventional hardening cement process disturbs both the kinetics and the mechanisms of cement hydration reactions, consequently interfering with early-age and latter-age performance and the durability of cementitious materials cured through carbonation.

As mentioned before, the carbonation reaction is responsible for the precipitation of $\text{C}\bar{\text{C}}$ in the porous space, which is the reason why a densification of the microstructure is generally accepted with alteration in the transport properties, namely, lower absorption and permeability of carbonated concretes [47,67,68]. However, some studies report a similar total porosity at later ages, namely, 28 and 90 days, in cementitious specimens with conventional and carbonating curing. Hence, the mechanism responsible for the decrease in the transport properties should be more supported on an alteration of the pore structure than on a reduction in total porosity [47,66,69,70]. In fact, at early ages, the carbonation reaction has a more effective impact on the reduction of large capillary pores, within the 50–100 nm range, while the hydration reaction tends to act on smaller pores, within the 5–10 nm range [77,78]. Thus, the coupled effect of both reactions at later ages is a microstructure with a lower number of pores larger than 50 nm when compared with a similar cementitious specimen with conventional curing [52,59]. This explains why various studies have shown that carbonation curing of cementitious materials hampered degradation mechanisms, namely, freeze–thaw, chloride penetration and carbonation-induced corrosion, consequently improving the material durability [51,78–80].

Following the carbonation stage, the post-conditioning stage is responsible for resetting the pH level of the liquid phase, increasing the alkalinity to values similar to conventional curing [52,54]. It is important to point out that the protection of rebars against corrosion is favoured due to the microstructure densification and high alkalinity level in the liquid phase, but it may be penalized by the slight reduction in overall carbonatable cementitious material [48].

Carbonation curing of cementitious materials potentially generates more reaction products, considering that both carbonation and hydration mechanisms are involved, and a cementitious matrix with reduced transport properties, when compared with conventional curing. In fact, the intermix between the hydration products, mainly C-S-H, and carbonation products, mainly $\text{C}\bar{\text{C}}$, promotes an increasing toughness and hardness, provided by the strong bonding between these two compounds [75,81]. Hence, carbonation curing of cementitious materials demonstrates an acceleration of strength development reactions and an increase in microstructure densification, compared with conventional curing. This early-age compressive strength increase is also found at later ages, being however usually negligible [48,52].

3.4. Impact on Carbon Capture

Estimates of CO_2 uptake in the carbonation curing process vary within a range, due to the many influencing factors present (e.g., carbonation conditions, which in turn depend on the chemical and microstructural characteristics of the material). Nevertheless, studies point out that a value between 20 and 80 kg of CO_2 per m^3 of concrete can be considered unanimous, which corresponds to about 7–27% of the total CO_2 emissions of concrete

(based on Figure 1 and Table 1). Moreover, besides absorbing CO₂, the process has been demonstrated to be less energy-consuming than steam curing, further contributing to the carbon emissions mitigation [51]. Additionally, the utilization of flue gas instead of pure CO₂, along with a pre-conditioning stage that takes advantage of natural environmental conditions, can improve the energetic cost of the process [17,48,82].

In summary, despite being applicable only to the precast concrete industry (accounting for 20–30% of the total concrete industry, including dry- and wet-mix products), the carbonation curing process represents a feasible prospect for CCUS technology, minimizing the CO₂ emissions associated with the cement industry.

4. Carbonation of Cementitious Materials during Mixing

In this strategy, CO₂ is applied during the mixing process before the casting and curing stages, providing a carbonating environment for the mixing of cementitious materials. This strategy addresses a stage prior to the one depicted above (the curing stage) and this aspect makes possible its wider application beyond the precast industry, extending its potential to in situ concrete. The theoretical concept behind this strategy is based on the following premises. By dispersing the cement particles in water, as occurs in this phase of the production process, CO₂ is able to fully access these particles and the auto-blocking effect from carbonate precipitation in porous space does not take place. Thus, two favourable conditions for carbonation occur (absent in carbonation curing): a more complete access of CO₂ to the carbon-reactive compounds; and absence of the porous solid barrier through which CO₂ has to diffuse. Following a structure similar to the one used for the previous strategy, the carbonation process, corresponding reactions and phase development, impact on microstructure, material performance and CO₂ uptake of this technique will be discussed in the following.

4.1. Carbonation Mixing Process

The conceptualization of this process comprises the utilization of CO₂ as a mixture component. Thus, all the reactive compounds that will produce the cementitious matrix, namely, cement, water and CO₂, are put into contact at the same time and mixed together. Diverse attempts were made to apply this conceptual idea. Kwasny et al. [83] and Silva et al. [84] exploited the utilization of mixing water previously carbonated in the mixing process. However, the expected carbon uptake will be small, given the low solubility of CO₂ in water at atmospheric pressure (0.0015 gCO₂/gH₂O [85]). Similarly, Nogueira et al. [86] and Kwasny et al. [83] opted instead to introduce the CO₂ mixture component in the atmosphere of an airtight chamber (the later study adopted both strategies simultaneously), using atmospheric pressure and a CO₂ concentration of around 80–90% *v/v*. Nevertheless, the most common process corresponds to the injection of CO₂ into the cementitious mixture during the mixing stage. In fact, this process has been used for a long time in the industry of wood-fibre boards to increase product turn-over [87]. Recently, CarbonCure Technology Inc. implemented a process that consists of injecting CO₂ directly into concrete while it is mixing in a truck. This injection lasts for 1–2 min and the amount of CO₂ injected is reported to be lower than 1.0 % by weight of cement [88].

There are still few authors working on this strategy, with each of them eventually exploring different processes and seeking to improve their efficiency [73,86,89,90]. Hence, a generic carbonation process has not yet been established, unlike the carbonation curing strategy. Moreover, these studies report some setbacks, stating a performance reduction for amounts of injected CO₂ above a given value. Therefore, controlling this parameter is crucial for the industrial viability of this strategy. One option already explored by the industry consists of dosing CO₂ in a flow-through or enclosed system for a given time period and respecting an optimal dosage, as presented in Table 2. In fact, the CO₂ injection system developed by CarbonCure Technology Inc. has returned improvements (after 3 days) of about 10% in the concrete's compressive strength after injecting a precise dosage of CO₂ during mixing [91,92]. This strength increase can be used to reduce the clinker

amount by 7 %, further reducing the CO₂ footprint. The concrete produced did not show flash setting nor workability problems. Other properties such as reduction in pH or density were also unaffected [88]. However, results are contradictory in the few studies published so far (Table 2), suggesting the need for further research. Mechanical performance greatly depends on carbonation conditions usually associated with low CO₂ uptake.

Table 2. Compressive strength improvement and footprint benefit of cement-based mixtures exposed to carbonation during mixing.

Mixture	Process	Compressive Strength Variation			CO ₂ Uptake	Refs.
Cement paste (w/b = 0.15)	Flow-through system Flow rate 5 L/min MD ¹ 240 s cCO ₂ ² 99.5%	<i>f_c</i> ³ variation −84% 3 days −83% 28 days			CO ₂ uptake 3.4%wt	[73]
Cement paste (w/b = 0.44)	Enclosed system cCO ₂ ² 85 ± 5%v/v MD ¹ 45 min MD ¹ 90 min	<i>f_c</i> ³ variation at 28 days −12% −13%			CO ₂ uptake 0.93%wt 1.12%wt	[86]
Ready-mix concrete (308 kg of cement and 77 kg of slag)	Enclosed system with injection of CO ₂ 0.05%wt 0.15%wt 0.30%wt (CO ₂ /cement)	No impact on durability <i>f_c</i> ³ variation 3 days 7 days 28 days			CO ₂ uptake	[92]
Ready-mix concrete (147.7 kg of cement and 73.9 kg of slag and of fly ash)	Enclosed system with injection of CO ₂ 0.11%wt (CO ₂ /cement)	Same <i>f_c</i> ³ as a reference concrete with 4.3% more binder			Reduction in CO ₂ emissions 10.0 kg/m ³ (CO ₂ /concrete)	[93]
Ready-mix concrete (25% fly ash)	Enclosed system with injection of CO ₂ 0.25%wt (CO ₂ /cement)	<i>f_c</i> ³ variation +24% 1 day +29% 2 days +23% 7 days +19% 28 days			N/A	[89]
Mortar (w/b = 0.5)	Enclosed system cCO ₂ ² 99.5%	<i>f_c</i> ³ variation 3 days 7 days 28 days			CO ₂ uptake	[90]
		−1.9% −9.6% −7.7%	−5.7% −7.1% −5.7%	−3.3% +5.6% +8.9%	0.44%wt 1.32%wt 2.20%wt	

¹ Mixing duration; ² CO₂ concentration of the contacting atmosphere; ³ Compressive strength.

Note that He et al. [73] applied carbonation curing (Table 1) and carbonation during mixing (Table 2) in mixtures with the same composition; however, they reported different results in both mechanical performance and CO₂ uptake. In fact, carbonation curing created a cement paste with a higher CO₂ uptake (7.3% versus 3.4%) and a much higher compressive strength (on average seven times higher throughout the hydration period) than in carbonation during mixing.

An alternative strategy consists of using mixing water previously carbonated, as depicted in Table 3. However, the CO₂ uptake achieved by this strategy was similar to that of the previous one in the few studies performed.

Table 3. Compressive strength improvement and CO₂ uptake of cement-based mixtures using carbonated mixing water.

Mixture	Compressive Strength Variation	CO ₂ Uptake	Refs.
Cement paste (w/b = 0.5)	f_c^1 at 28 days 6% reduction	N/A	[84]
Cement paste (w/b = 0.4)	f_c^1 at 28 days 20% reduction	N/A	[94]
Cement mortar (w/b = 0.4)	Reduction in f_c^1 Porosity doubled and pores around 80 nm increased	N/A	[94]
Cement paste (12% limestone filler) (w/b = 0.4)	f_c^1 variation at 1 day +18%	0.61%wt	[95]
Mortar a/c = 0.4	f_c^1 variation 1 day 3 days 7 days −9% +18% +13%	1.03%wt	[96]

¹ f_c —compressive strength.

4.2. Reactions and Phase Development

Carbonation of anhydrous and hydrated compounds, following Equations (5)–(7), does take place but the previously reported low value of CO₂ uptake suggests that these carbonation reactions are negligible. Nevertheless, the different performance obtained in the cement paste, either in fresh or in hardened state, suggests that the carbonation at this early stage interferes in the ordinary hydration reactions of cement. The chemical nature of the reactions and newly arising compounds may be the same but the kinetics of the reactions and the final paste microstructure are different [83,86,91].

Cement paste exposed to CO₂, above a certain threshold, presents a high hydration heat and flash false setting that punishes workability [83,86,87,97]. This increase in consistency occurs immediately after the addition of water and CO₂. However, after an initial peak of hydration heat, the mixture goes into the induction period, more or less at the same time as the reference non-carbonated paste [86,95]. Moreover, the acceleration period is longer, the hydration heat shows a slower increase and the maximum peak is also lower. Hence, CO₂ seems to have a set acceleration effect in the pre-induction period and a set retardation effect in the acceleration period [86]. However, for low amounts of CO₂, only the set acceleration effect is visible [91,95]. This behaviour has been previously observed with other admixtures, namely, those based on triethanolamine (TEA) and on carbonates such K₂CO₃ and Na₂CO₃ [87,98,99]. This small adjustment in admixture amounts causes such changes in events that lead to a product with much better performance [92].

Analysis of the evolution of carbonation and hydration reactions shows that \overline{CC} arises immediately after CO₂ comes into contact with cement and water. However, \overline{CC} is generated in the form of nanoparticles of amorphous or poorly crystalline nature. These particles appear along with fibrous particles (attributed to C-S-H) and etch pits on anhydrous grains [91]. This high activity at this early stage contrasts with the dormancy that occurs in reference pastes, where almost no product formation and no pitting is visible. This suggests a more effective dissolution of anhydrous phases, which will be the cause for the argued set acceleration. Yet, this activity is of short term, resulting in a longer induction followed by a more extended acceleration. The actual cause for the end of the induction period has not yet been demonstrated but there is a systematic coincidence with the precipitation of portlandite [100], although not necessarily with a corresponding undersaturation with respect to the solutes [101]. In fact, the first CH products to appear are amorphous highly hydrated compounds, which apparently contribute to raise the maximum concentration of calcium ions in solution at about twice the solubility of portlandite [100,102]. These compounds evolve through different species until they become well-crystallized portlandite. \overline{CC} also

shows a similar behaviour, evolving from a highly hydrated amorphous structure that eventually ends up as calcite [103]. In fact, many works on carbonation strategies of cementitious materials mention the higher amount of amorphous CH on the first day, in relation to the reference uncarbonated paste [91,95,97]. Hence, the mechanism behind the induction is argued to be similar, whether with or without CO₂, both being based on the capacity of the solution to contain a calcium concentration above the portlandite or calcite saturation degree (only portlandite in reference paste, both calcite or portlandite in paste with CO₂) [86]. A higher amount of amorphous CH was also observed in mixtures exposed to CO₂, in relation to the reference. Moreover, the rate of portlandite precipitation and growth is slower and the process takes longer, which has been pointed out as the cause of the flat acceleration of carbonated cement pastes [86].

4.3. Carbonation Impact on Microstructure and Material Performance

The main problems of cement mixture performance include flash false setting that hampers workability during the fresh state and reduction of compressive strength during the hardened state; as mentioned before, both are related to the introduction of CO₂ above a specific threshold [84,86,92]. In fact, the oversupply of CO₂ causes a looser and more disconnected microstructure compared with the reference paste. Nogueira et al. [86] theorized that the CO₂ produces a heterogeneous paste with larger CH crystals intercalated with C-S-H, which could justify the reduction in compressive strength [102,104]. Additionally, the coarsening of the pore structure from a unimodal distribution of around 0.1 µm to a widened distribution from 0.1 µm to 1 µm, while keeping the same total porosity, further supports this performance reduction [86].

4.4. Impact on Carbon Capture

The negative impact of CO₂ above a specific threshold on the early hydration of cement, greatly reduces the carbon uptake potential of cementitious materials exposed to carbonation during the mixing process.

Only for CO₂ dosages below about 1% of cement weight is it possible to prevent the described negative effects on material performance. For this small dosage, it is possible to control the set-accelerator effect of CO₂ on the hydration reactions, thus avoiding the flash false setting and increasing the early mechanical strength. This mechanical strength increase can be offset by cement reduction, which eventually leads to a higher CO₂ reduction in terms of unit volume of concrete. Taking this into account, Monkman et al. [88,92] reported 14.8 kg of CO₂ saved per cubic meter of concrete. Nonetheless, the information presented in Table 2 suggests a more modest impact of this strategy on the mitigation of CO₂ emissions: on average 3.3 kg of CO₂ per m³ of concrete, assuming a maximum CO₂ absorption of 1% of cement weight and disregarding the possible reduction in cement consumption.

5. Carbonation of Hydrated Cement Waste from Demolished Concrete

The construction industry sector is responsible for the production of about 450 million tonnes of construction and demolition waste (CDW) per year, accounting for 35% of the total waste stream generation in the European Union [105,106]. Considering the diversity of construction and demolition activities, it is estimated that only 12–40% of CDW is composed of concrete [107,108]. Dumping and landfilling of CDW is currently the main waste management approach undertaken, contaminating soil and groundwater with the hazardous substances present in concrete and further increasing the environmental footprint of the construction sector. To overcome this negative impact, technologies focused on the recycling of CDW have been increasingly developed. The most viable method for CDW disposal, namely, end-of-life concrete, is its conversion into recycled concrete aggregates (RCA) [109,110]. This recycling process generates fines very rich in cement, corresponding to the binding paste adherent to the aggregates that separates during the grinding of concrete waste. These fines are commonly referred to as hydrated cement waste (HCW) and are considered a by-product of the recycling operation [111–113].

Diverse studies have explored the application of HCW in cement-based materials, namely, as supplementary cementitious material (SCM) [114–118]. The reported results indicated that the positive performance of a cement mortar with HCW addition was limited to 5% replacement. With the same purpose, other papers addressed the carbonation of HCW prior to its application as SCM. Lu et al. [111] investigated the impact of adopting fully carbonated HCW (CHCW) obtained from pure cement paste and uncarbonated HCW as binder additions on the compressive strength (f_c) of cement pastes. In this study, the replacement of cement with CHCW produced a performance increase of 6% and 32% for 10% and 20% incorporation, respectively, in relation to the same incorporation values of HCW. Also, Mehdizadeh et al. [119] reported similar results: the incorporation of 5–20% of CHCW returned a positive increase in f_c when compared with a reference paste composed of 100% cement. Thus, a feasible prospect for a CO₂ capture technology was uncovered [120,121]. In short, this strategy consists of exposing HCW to CO₂, creating a material with composition and fineness similar to limestone filler, which is commonly used throughout the world in concrete production. Therefore, this carbonated material can be utilized as an addition in new concrete batches as an alternative to limestone filler.

The alkali activation [122,123] and the thermoactivation of HCW [124–126] have also been explored in an attempt to improve the HCW performance as a binder addition, although discarding the CO₂ uptake technology; these aspects are beyond the scope of this text.

5.1. Carbonation of Hydrated Cement Waste

Despite the positive results obtained by carbonated HCW as an SCM, the process required to achieve a high-enough carbonation degree still needs optimization. The carbonation process is usually very long (from 12 to 28 days), with environmental conditions of RH = 60–70%, temperature of about 20 °C and CO₂ concentration from 20 to 100% [81,119,127]. In contrast, Wu et al. [128] set a carbonation duration of 3 days, remarking that HCW presents a rapid reaction with the CO₂ and that, after this initial carbonation rate boost, the carbonation efficiency slows down significantly. Recently, Silva et al. [129] investigated the carbonation mechanism and kinetics of HCW during a short duration carbonation process of two hours, reporting a rapid carbonation in the first 20 min, followed by a progressive reduction in the carbonation rate. Moreover, the results further indicate that diverse calcium-bearing components of HCW are involved in the reactions during the initial rapid carbonation rate and, in the following slow rate period, only C-S-H appears to contribute to the carbonation reaction. In fact, the carbonation behaviour of CH and C-S-H, the two main calcium-bearing cementitious compounds, is very distinctive throughout the carbonation process; while CH reached a carbonation degree of 85% after 20 min, attaining almost full carbonation, C-S-H achieved a carbonation degree of only 30% in the first 20 min, and 32% at the end of the process [129,130]. Nevertheless, the absence of further investigations focused specifically on the carbonation process hinders extensive analysis of the parameters with potential to optimize this process.

Another approach also explored by academics is the impact of some HCW physical characteristics on the carbonation process, namely, its particle dimension [131], the water/binder (w/b) of the original hardened paste waste [127] and HCW purity [131]. Mehdizadeh et al. [127] reported a higher CO₂ uptake of HCW from cement pastes with higher w/b. The CO₂ uptake increased by about 15% when the w/b was increased from 0.2 to 0.4. Silva et al. [131] reported that the CO₂ uptake increased by only 1.3% when the HCW maximum particle size decreased from 250 µm to 75 µm. However, the HCW initial water content revealed a great impact, varying the CO₂ uptake from 6% to 29%. Regarding the HCW purity, two strategies are usually adopted to overcome the variability and complexity of HCW composition due to the diverse origins of end-of-life concrete collected from different demolition sites. Early studies were compelled to resort to synthetic laboratory-made cement pastes with controlled composition. Typically, synthetic HCW is produced by crushing and sieving of hardened pastes (after a curing period of about 28–90 days)

into a particle size usually below 150 μm . More recent works sometimes opt to remove the aggregate impurities existing in industrial HCW. In this context, Carriço et al. [132] developed a novel separation method that allows the retrieval of HCW from concrete waste with nearly 90%vol purity. This makes it possible to increase the absolute volume of CO_2 absorption of HCW per cubic meter of treated material. However, Silva et al. [131] showed that the carbonation rate of HCW produced under synthetic lab-made conditions and that made from end-life-concrete is similar, producing similar carbonated cement pastes with comparable carbonation degrees, further indicating that the presence of aggregate impurities in HCW does not hinder the progress of carbonation.

The environmental parameters relevant to the carbonation setup have also been analysed [131,133]. Conclusions were similar to those obtained for carbonation curing (Section 3.1). In general, an increase in the severity of carbonation conditions (e.g., CO_2 concentration and relative pressure of the carbonating atmosphere) is favourable to the CO_2 uptake until a defined threshold, where the precipitation of carbonate compounds hampers the CO_2 diffusion, blocking the CO_2 absorption [129,131]. Table 4 presents the CO_2 uptake and compressive strength improvement reported in some recent works on the carbonation of hydrated cement waste from recycled concrete.

Table 4. Performance and CO_2 uptake of cement mixtures containing CHCW.

HCW Origin	HCW Conditions ¹	Carbonation Process ²	CO_2 Uptake (%wt of HCW)	Mixture	Compressive Strength Variation			Refs.
					1 day	28 days		
Lab-made paste (w/b = 0.4)	<75 μm	N/A c CO_2 99% 1 atm 20 °C RH 60%	20.9	10% ³	+16%	+4%	[111]	
				20% ³	+24%	+12%		
				30% ³	4%	−5%		
				Cement paste (w/b = 0.4)				
Lab-made paste (w/b = 0.3)	0–75 μm 75–150 μm	28 days c CO_2 20% 1 atm 20 °C RH 65%	21.1	10% ³	+5%	0%	[119]	
				20% ³	+8%	0%		
				30% ³	0%	−3%		
				Cement paste (w/b = 0.3)	75 μm	150 μm		
Lab-made paste (w/b = 0.3)	<200 μm	12 days c CO_2 20% 1 atm 20 °C RH 70%	19.3	30% ³		N/A	[81]	
				Cement paste (w/b = 0.4)				
Lab-made paste (w/b = 0.2) (w/b = 0.3) (w/b = 0.4)	<75 μm	28 days c CO_2 20% 1 atm 20 °C RH 65%	18.3 for 0.2 19.7 for 0.3 21.0 for 0.4	5% ³	+4%	−1%	0%	[127]
				10% ³	+7%	+7%	+5%	
				15% ³	+8%	+10%	+6%	
				20% ³	−6%	−2%	+9%	
				Cement paste (w/b = 0.3)	0.2	0.3	0.4	
Lab-made paste (w/b = 0.45)	<250 μm WC 17%	2 h c CO_2 80% 1 atm 20 °C RH 70%	19	N/A		N/A	[129]	

¹ maximum particle size; ² duration/ CO_2 concentration/ CO_2 pressure/temperature/RH; ³ percentage of cement replacement.

5.2. Reactions and Phase Development

As HCW is a material obtained from concrete at the end of its life cycle, it is composed of highly hydrated compounds, unlike the anhydrous or partly hydrated cement compounds found during concrete mixing and curing, respectively. Similarly, the presence of carbonates is also higher in HCW due to natural carbonation. In other words, in previous carbonation technologies the cement compounds are in an early stage of hydration, while in this technology, hydration is almost complete. Hence, HCW is a calcium-rich material containing mainly CH and C-S-H, with a negligible quantity of anhydrous compounds [48,110,134,135].

During the carbonation process of HCW, both CH and C-S-H produce \overline{CC} . The carbonation of CH is a simple process in which CO_2 reacts with CH producing H_2O and \overline{CC} , following Equation (2). Conversely, the carbonation process of C-S-H is more complex, characterized by a decalcification and polymerization of its structure with formation of a modified C-S-H with a lower Ca/Si ratio and, eventually, silica gel (S-H) (Equation (6)). Regarding the \overline{CC} produced, studies report the formation of different polymorphs [53,129,136]. These compounds may arise in a highly interconnected manner, namely, as a composite formed by intermixing of decalcified C-S-H and amorphous \overline{CC} phase [53,81]. For extended carbonation, amorphous highly polymerized S-H may arise [137]. Moreover, the carbonation process is described as occurring simultaneously for both compounds (CH and C-S-H) [129,138,139], with the carbonation degree being increasingly favoured for aged cement compounds like HCW, in comparison with early-aged ones [140]. Thus, considering previous studies [55,133] and Equations (6) and (7), Figure 4 presents an estimation of the composition of HCW (left) and HCW with calcium oxide content fully carbonated (right), illustrating the main phase transformation due to the full carbonation of HCW.

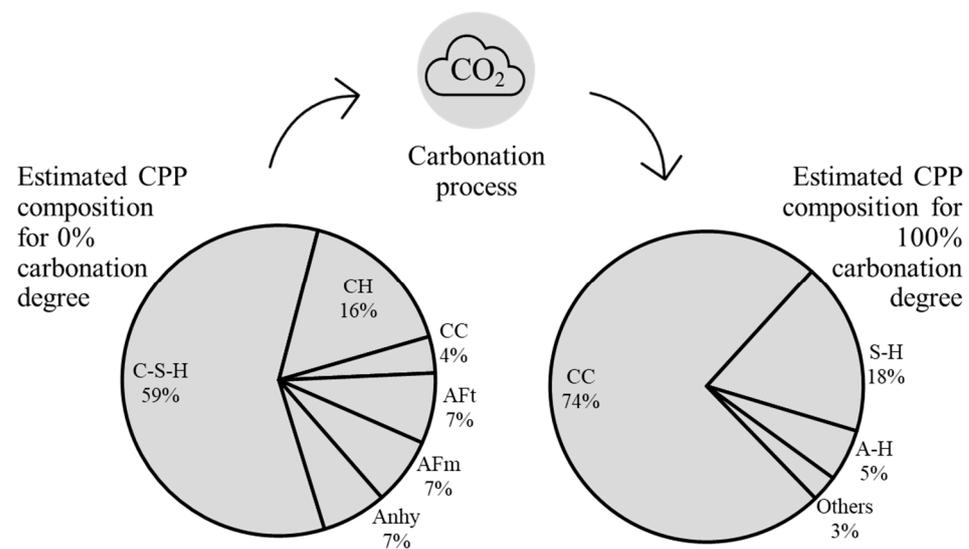


Figure 4. Main phases in HCW (left) and after the carbonation process (right). Legend: anhydrous cement (Anhy); calcium silicate hydrate gel (C-S-H); silica gel (S-H); calcium hydroxide (CH); ettringite (AFt); monosulfate (AFm); alumina hydroxide (A-H) [55,129,133,141,142].

5.3. Carbonation Impact on Microstructure and Performance

Studies addressing the potential of CHCW as an SCM have mainly focused on the performance characteristics evaluated in mortars and pastes. The generic long carbonation process usually applied in different studies alters the initial HCW composition, as aforementioned, creating a CHCW with better characteristics for SCM. In fact, different studies analysed an extensive and continuous set of replacement percentages of cement with CHCW and HCW, reporting a higher compressive strength for the utilization of CHCW in every replacement percentage (from 5% to 30% wt.% of binder). Moreover,

for replacement percentages below 20%, the utilization of CHCW as an SCM produced a compressive strength either similar to or higher than that of the reference cement paste with 100% of cement, contrarily to the HCW utilization, which consistently produced a lower compressive strength [111,119,128]. Furthermore, Wu et al. [128] pointed out similar water transport properties between cement mortars with the addition of CHCW and the reference mortar with 100% cement.

The positive contribution of CHCW is due to the CC formed in the carbonation process. As argued for limestone additions in blended cements, the presence of CC in CHCW promotes a nucleation effect, responsible for the increase in growth sites for the cement hydration products and for the spacing of anhydrous grains, preventing the growth blockage of those products [111,119,128]. Simultaneously, the stabilized calcium carbonate present in the CHCW addition may react with the aluminate phase of cement, allowing the formation of calcium aluminate monocarbonate (Mc) and calcium aluminate hemihydrate (Hc), instead of the mono-sulphoaluminate phase (Ms). Hence, the overall impact of adopting CHCW as SCM includes mechanical strength increase and porosity reduction in relation to the reference paste with only cement, provided the incorporation is kept below a certain amount. A lower water demand was also identified in CHCW-added mixtures when compared with those using HCW [48,109,110].

Furthermore, the particle size influence of HCW was also investigated by Mehdizadeh et al. [119], who reported an increase in the compressive strength of mortars with CHCW particles smaller than 75 μm when compared with those with particles between 75 μm and 150 μm . The authors attributed this behaviour to the greater nucleation effect and the faster formation of Mc (higher reactivity). Mechanical strength improvement was also reported in HCW (without carbonation) subjected to longer grinding periods [110].

5.4. Impact on Carbon Capture

The environmental impact of CHCW as an SCM in the concrete industry is still unexplored, a consequence of the novelty and early development stage of this carbon capture strategy. Nevertheless, it is possible to estimate the carbon uptake generated by this technology from values reported in related investigations performed by the scientific community. Van der Zee et al. [143] estimated that 0.056 tonnes of CO_2 can be sequestered by each tonne of concrete waste, assuming 13–18 %wt of cement in concrete. More recently, studies have disclosed that fully carbonated CHCW is able to uptake 0.24–0.28 g of CO_2/g , which, considering an average of 15% cement in concrete waste, corresponds to 0.036–0.042 g of CO_2/g of concrete waste [81,111,133,134]. This uptake can be improved if more pure hydrated cement waste is previously retrieved from concrete waste. These estimates assume the same carbonation degree as achieved in lab experiments, which in general corresponds to complete CH carbonation and incomplete C-S-H carbonation.

Moreover, the use of CHCW as an SCM reduces the amount of clinker, further contributing to the reduction of CO_2 emissions beyond the carbon capture previously addressed [115]. Hence, considering an average net emission of 292 kg of CO_2 per m^3 of concrete [2,9,10,133], and a 20% replacement of clinker by CHCW, this strategy is capable of reducing clinker-related CO_2 emissions to 221 kg of CO_2 per m^3 of concrete, a reduction of 24% (20% from cement substitution and 4% from CO_2 absorption by CHCW).

6. Overview

The three CCUS technologies discussed in this paper were chosen based on the recognition they have received from academia and industry in the last decade, especially the carbonation curing strategy, and also based on the high marketability potential which also drives continuous investigation of carbonation during mixing and carbonation of hydrated cement waste from demolished concrete.

The first strategy, carbonation curing, is undoubtedly the most investigated one. A number of studies have addressed specific concrete products and experimentally obtained CO_2 uptake values ranging, on average, from 20 to 80 kg per m^3 of concrete [144]. However,

this value only considers the CO₂ captured by cement compounds and does not indicate a net CO₂ benefit. In fact, the most challenging issue of this strategy is to establish an industrially viable process by considering the CO₂ life cycle of all the upstream processes, including CO₂ capture, transport and utilization, as well as the potential net CO₂ benefit from any improvement in mechanical strength. A literature review based on 70 carbonated curing concretes showed that only 26% had positive net CO₂ [144]. But these results can change depending on the assumed premises, which are sometimes highly controversial. For instance, the source of the consumed electricity (fossil fuel, natural gas or renewable sources) or the source of the CO₂ (power plant or cement plant, location near or far from the carbonation facilities) in cases other than the CO₂ waste scenario, for which a viable outlet is required, are matters of debate. A previous review showed that the compressive strength of concrete products at 28 days decreased in 27% of the cases, although their early compressive strength improved [144]. Moreover, this strategy still has some unresolved issues, essentially related to CO₂ emissions from the high electricity consumption of the curing process. The adoption of a lower CO₂ content atmosphere for curing is another possibility that should be explored. In addition, one should keep in mind that carbonation curing is limited to the precast concrete industry (around 20–30% of total concrete production) and to some prefabricated large elements, due to practical reasons.

Carbonation during mixing, the second strategy referred to in this paper, has some pros in relation to carbonation curing, namely, the higher exposition of cement compounds to CO₂, since they are in a powdered form and in suspension in a CO₂ solution. The industrial process has the potential to be simpler and less energy-consuming, as only the carbonation stage is required (instead of pre-conditioning, carbonation and post-conditioning). According to Ravikumar et al. [144], the net CO₂ benefit was positive in 86% of the 29 published cases studied. Moreover, this strategy has a broader target market, the entire cement-based materials industry. However, the introduction of CO₂ in the mixing stage, where the cement hydration reactions are still very incipient, may cause a very detrimental impact on the evolution of those reactions, affecting the mechanical and durability performance. To avoid this issue, CO₂ has to be added in a very low amount (less than 1 %, in terms of mass of cement, which is 3 kg per m³ of concrete if 300 kg of cement per cubic meter of concrete is assumed), wherein a positive accelerator effect is visible. Monkman et al. estimated a net CO₂ benefit of 14.8 kg of CO₂ per cubic meter of concrete, taking into account the clinker reduction from strength increase and the slight CO₂ absorption from the carbonation process [88,92]. Therefore, the potential of this strategy relies on the high volume of the target market.

Finally, in the third strategy, the valorization of hydrated cement waste from demolished concrete structures has the important advantage of promoting the recycling of a by-product without a clear outlet, rather than interfering with the established industrial processes previously mentioned. Abundant amounts of hydrated cement waste are currently available and will be in the near future, considering the lowest estimates of 54–180 million tonnes per year of concrete waste produced in the European Union alone. For an industrial implementation, the waste grinding and carbonation processes (ensuring suitable CO₂ partial pressure, RH and temperature) were identified as the main energy consumers. Studies on the overall net CO₂ impact are unknown, since research on this strategy is scarce, but estimates point to an uptake of 240–280 kg of CO₂ per tonne of hydrated cement waste, returning the significant amount of about 40 kg of CO₂ per cubic meter of concrete waste. However, it is important to note that these values should be corrected downwards by the amount of CO₂ emissions from the upstream processes (namely, waste grinding and transport and electricity consumption of the carbonation process). Conversely, the obtained product can also reduce CO₂ emissions if it has a viable utilization, namely, as a cement replacement, as seems to be the case. In addition, if concrete waste separation is carried out, the concentration of cement will rise and the whole process becomes more operational. The most promising results suggest strength increments between 6 and 32% for a carbonated hydrated cement waste (CHCW) incorporation of 10–20%. Experimental work to model

the overall CO₂ cycle and determine the net CO₂ value also needs to be developed because the balance between CO₂ emissions and capture is very sensitive to small adjustments in the process. For instance, an intensification of waste grinding returns benefits in terms of the mechanical performance of the final mixture, optimizing the utilization potential of HCW as an SCM; the optimization of the CO₂ uptake by HCW has to be considered in light of the carbonation process efficiency. In addition, contrary to other strategies, the carbonation of hydrated cement waste also promotes the reuse of CDW and the saving of natural resources, further contributing to the reduction of the environmental impact of concrete. The pros and cons of the three strategies are summarized in Figure 5 to assist further understanding of the carbon mitigation potential of each strategy.

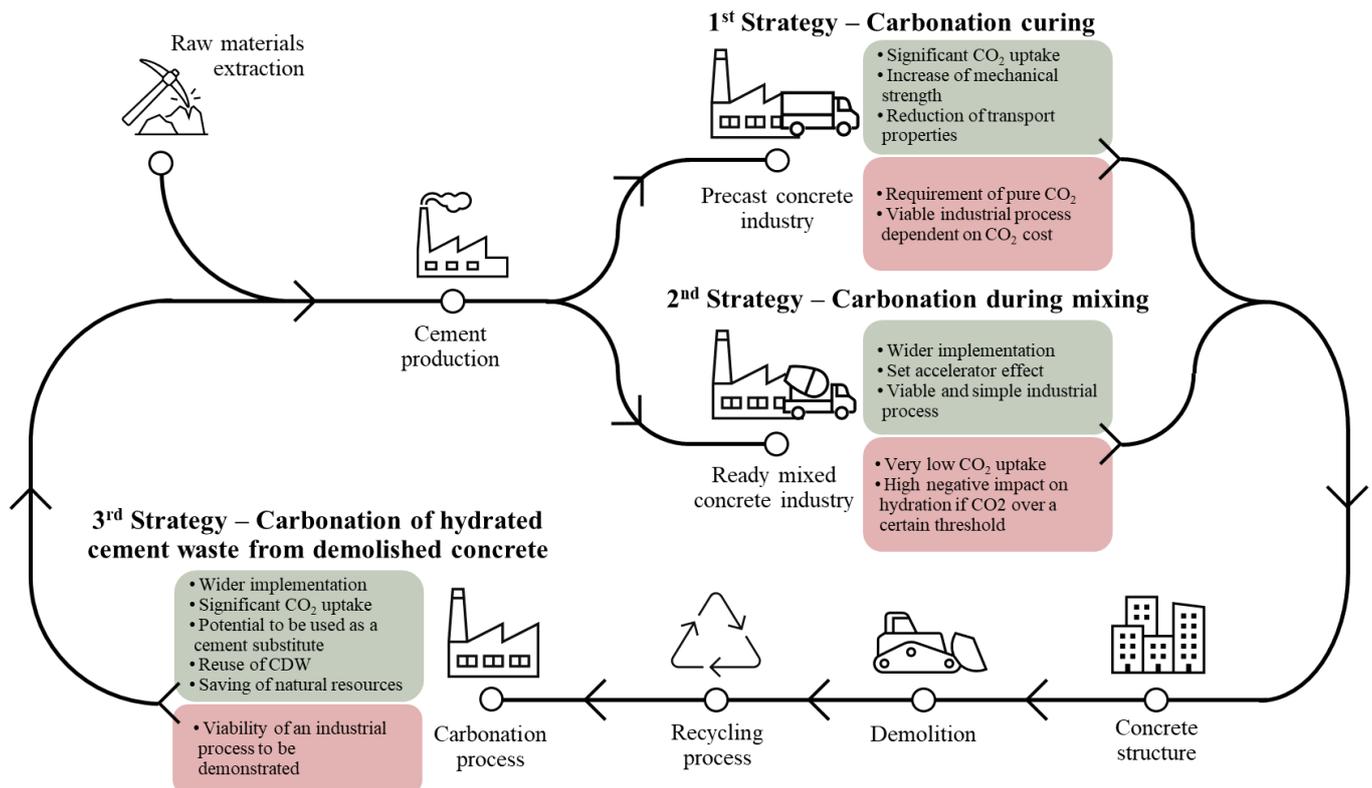


Figure 5. Pros and cons of the CCUS strategies.

Figure 6 presents a quantitative comparison of CO₂ uptake between the CCUS strategies, illustrating the corresponding contribution to the reduction of these emissions. This analysis considers a CO₂ uptake for each strategy, based on the values collected from the literature and previously presented in Tables 1, 2 and 4. Therefore, a CO₂ uptake of 14%, 1% and 20% (in terms of the cement mass) was considered for carbonation curing, carbonation mixing and carbonation of HCW, respectively. An amount of 328 kg of cement per m³ of concrete was assumed, both for the new productions (curing and mixing), as well as for the concrete from which HCW is retrieved and a high separation efficiency was considered in this last case. Moreover, the calculations for the third strategy assumed that the carbonated HCW was used as a binding material, replacing 20% of the cement amount in the concrete production. This assumption leads to a reduction of 24% of the CO₂ emissions, also resulting from the lower cement consumption. Then, for a proper comparison of the strategies, a similar replacement of cement is considered for the first two. Here, besides the carbonated addition, a replacement with an inert addition like the commercially available limestone filler was also considered. These cases illustrate the cumulative effect of using a carbonated addition (or an inert addition) in the production of concrete subjected to carbonation curing or carbonation mixing. The secondary CO₂-emitting processes (e.g., CO₂ transport and stor-

age) were disregarded in the analysis, thus providing a preliminary perspective on which strategy presents a better prospect. Furthermore, the consideration of these secondary processes would require specific research tasks outside the scope of this paper.

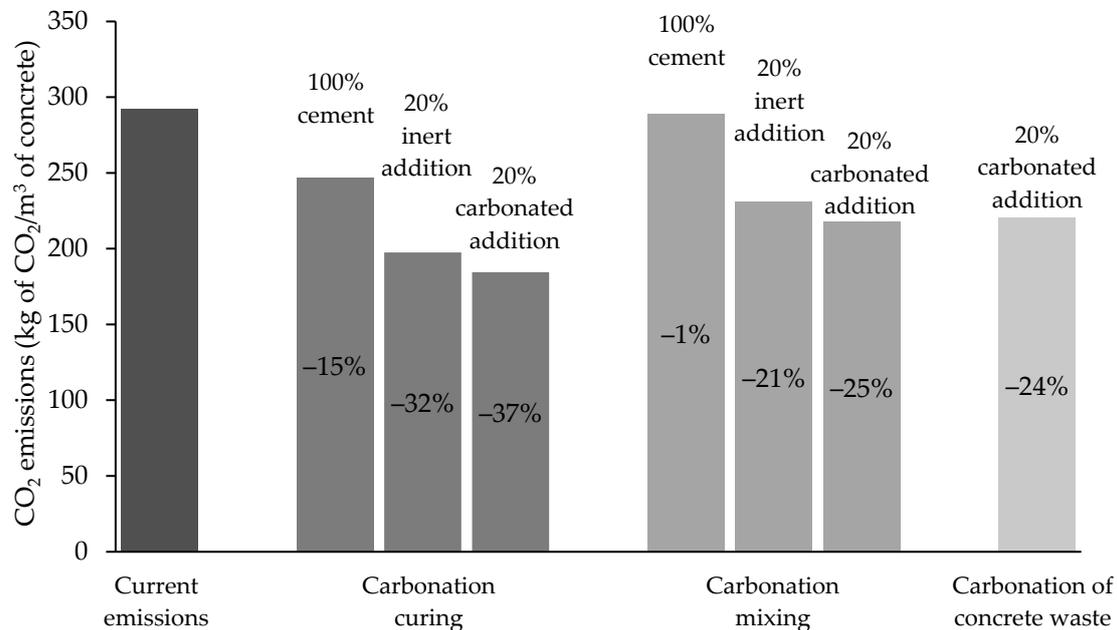


Figure 6. Comparison of the CO₂ uptake between the CCUS strategies.

Figure 6 shows that the biggest benefit occurs when the cement consumption is reduced. However, if the replacement consists of a carbonated instead of an inert addition, the CO₂ emission reduces by 4–5% more. The highest effect occurs from the adoption of a carbonated addition to produce a concrete undergoing carbonation curing, that is, a combination of the first and third strategies. In this case, the CO₂ emissions reduce from 32% to 37%, which is a significant increment in the CO₂ uptake, uncovering a promising prospect for the CO₂ emission mitigation target. It is important to note that the carbonation mixing strategy can also be used together with the other two, increasing the CO₂ uptake by 1% more.

These results point out that the maximum carbon emission reduction is around 184 kg of CO₂ per cubic meter of concrete when considering carbonation curing and carbonation of concrete waste together. Note that this value is an optimistic estimation since CO₂ emissions from secondary processes were disregarded. This estimation increases to 218 kg of CO₂ per cubic meter of concrete if considering only the strategies available for the ready-mix concrete industry. Both these results demonstrate that, besides the combination of CCUS strategies and the increase in their carbonation efficiency, more carbon mitigation technologies are required to achieve a carbon-neutral concrete product, such as a less cement-intensive concrete mix design, an addition with a higher CO₂ storage amount or a less carbon-intensive cement.

7. Conclusions

This paper discusses the carbonation of cementitious materials using different strategies and different stages of the concrete life cycle, providing some important conclusions.

The carbonation curing strategy presents a high CO₂ uptake, between 20 and 80 kg per m³ of concrete, although its application is limited to the precast industry, with a market volume of 20–30% of the whole concrete industry. Despite extensive investigation, the industrial viability of carbonation curing has yet to be demonstrated, essentially due to the high CO₂ costs, including its purification and other energy-demanding processes. This problem can be minimized by considering a wider carbon chain, integrating diverse carbon

dioxide utilization technologies. Thus, the main obstacle of this strategy lies in the improvement of the carbonation process efficiency, whose main challenges are the complexity of the curing procedure, composed of diverse stages, and the risk of compromising the mechanical performance and CO₂ uptake. Carbonation during mixing presents the lowest CO₂ uptake (<3–4 kg/m³) of the addressed strategies; however, the wider application field (theoretically, the generality of the cementitious materials) further promotes its carbon mitigation potential. Moreover, the ease of process implementation in the established system of concrete plants helps explain the already-existing commercial application of this strategy. Once again, the carbonation process efficiency is one main development focus of this strategy; thus, the optimization of the mixture (namely, by partially adopting SCM) or the utilization of flue gas from a nearby cement plant are approaches with good potential for increasing carbonation efficiency. The final strategy, the carbonation of cement waste from demolished concrete, presents a potential CO₂ uptake of around 40 kg per m³ of concrete waste; however, the research on this subject is still scarce, and its industrial viability is yet to be demonstrated. Contrarily to other covered strategies whose development is essentially focused on the process efficiency, this strategy should focus primarily on the effectiveness of carbonation. This may be achieved essentially by identifying the optimal carbonation conditions, since carbonation of all the calcium-bearing compounds is theoretically feasible.

Finally, it is important to remark that to achieve the zero-emissions target, the simultaneous implementation of different strategies is paramount as previously demonstrated, especially because they do not compete with each other. Therefore, despite the different CO₂ uptake potential and industrial feasibility of the strategies, they should be equally developed with the aim of introducing more than one CCUS strategy in the life cycle of concrete, together with other carbon mitigation technologies.

In future research, a stochastic analysis should be performed, focusing on a carbon mitigation capability comparison of the three strategies presented in this paper and considering the CO₂ emissions of secondary processes required for the application of each strategy. Furthermore, as different concrete products used in the construction industry present different performance requirements, which results in specific concrete industry sectors (e.g., ready-mix industry and precast industry) with distinctive capabilities to accommodate disruptive and new carbon mitigation technologies, it would also be important to comprehend which specific concrete products have the theoretical capability to achieve carbon neutrality, e.g., concrete blocks for masonry; thus, it would become possible to point out a clear research pathway for a more sustainable construction material industry.

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