Review

Carbonation Potential of Cementitious Structures in Service and Post-Demolition: A Review

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Abstract: The construction sector is responsible for a great environmental impact. The cement industry, which is included in this sector, emits about 650 to 800 kg of CO2 per each tonne of cement produced, being one of the most polluting industries in terms of greenhouse gas emissions. The cement manufacturing process releases about 7% of the total worldwide CO2 emissions. However, concrete and cement-based materials present CO2 uptake potential during their service life and post-demolition through carbonation processes. The carbonation reactions rate depends on several factors, namely type and content of cement, porosity of concrete, temperature, relative humidity and exposure conditions area. Therefore, to estimate the CO2 capture of concrete during its life cycle is not a straightforward calculation. Some studies have been developed using different methodologies in order to evaluate the CO2 potential of cementitious elements in service and post-demolition. This paper reviews the documented approaches that quantify the CO2 uptake of concrete over time, summarizing the assumptions adopted for each previous work. Overall, it was concluded that part of the CO2 emissions released during cement production are reabsorbed by concrete products during their life cycle, which partially offsets the environmental impact and reduces the CO2 footprint of the cement industry.

Keywords: carbonation; carbon dioxide uptake; cement industry; concrete elements; CO2 life cycle

1. Introduction

The cement industry is responsible for about 7% of the total worldwide CO2 emissions [1–3], mainly from the burning of fossil fuels and calcination of limestone for the manufacture of clinker. In summary, the production of 1 kg of Portland cement generates approximately 0.8 kg of CO2 [2]. Therefore, the search for alternatives to reduce greenhouse gas emissions is essential. It is necessary to improve cement production to avoid the emission of gases into the atmosphere and to reduce other types of environmental impacts. Mortars and concrete can capture CO2 over time through carbonation; thus, this process tends to partially compensate for CO2 emissions from cement production [4]. Usually, cementitious materials in service are exposed to water, which affects their characteristics over time. The CO2 present in the water can also accelerate reactions with calcium silicate hydrate gel, namely C-S-H, increasing the formation of CaCO3 [5,6]. It is well known that the concrete exposed to CO2 in aqueous solution may carbonate faster when exposed to the air.

Carbon dioxide sequestration rate of cementitious materials depends on several factors. The increment of this rate is responsible for a more carbon neutral concrete structure. Therefore, the carbonation of the cementitious structures in service must be investigated in order to update their life cycle in terms of carbon emissions [7]. This way, even though the production of Portland cement generates carbon dioxide emissions due to the energy usage and calcination process, a significant amount of CO2 can be re-absorbed through carbonation reactions [8,9]; bearing in mind that this process occurs very slowly, and usually
takes decades to reach significant levels. The carbonation reactions (responsible for CO₂ absorption) of cement-based products have not been deeply reported and consequently have not been considered in the CO₂ emissions assessment of cement production. Therefore, CO₂ emissions generated by cement factories can be overestimated [8]. In order to calculate the amount of CO₂ uptake from the atmosphere through carbonation processes by building materials, several studies have been carried out to estimate the proportion of CO₂ emitted by cement production which is re-absorbed over the years. This paper encompasses previous research that analyses carbonation during the service life of concrete structures as well as after demolition, namely in the form of recycled aggregates. This review is a summary of studies that have quantified the CO₂ uptake of cementitious structures over the years. However, each quantification of large-scale CO₂ uptake flux is based on several assumptions which allow these calculations.

2. Carbonation Reaction in Concrete

The carbonation process in concrete and mortars is the reaction between the calcium hydroxide (Ca(OH)_2) present in the cementitious matrices and carbon dioxide (CO₂). CO₂ ingresses into concrete through its pores. The cement hydration products in contact with CO₂ form CaCO₃ [10,11], changing the physical and chemical properties of concrete [5,12,13]. In the carbonation reaction, Ca(OH)_2 is consumed and Ca²⁺ and hydroxyl ions are released. The CaCO₃ content increases as long as there is availability of portlandite and calcium-silicate hydrates to react with CO₂ [14].

The propagation of CO₂ into concrete is a complex mechanism. After the development of a thin layer of carbonated concrete on the surface exposed to the atmosphere, the further propagation of CO₂ is hindered by the less porous carbonated layer. Therefore, CO₂ propagation does not occur at a constant rate, since concrete is a heterogeneous material and also due to the densified carbonated surface zone, which becomes thicker over time. Therefore, the carbonation depth increases linearly with the square root of reaction time, since the cementitious products carbonation rate is influenced by CO₂ diffusion [14–16].

Cementitious matrices capture CO₂ throughout their life cycle. Several factors influence the propagation of CO₂ into the concrete. The main aspects are the moisture content and the permeability of concrete, the content of pozzolanic material, the CO₂ concentration and the time of exposure [17–20]. CO₂ uptake capacity of cementitious materials depends on internal and external factors. Internal characteristics refer to the cement type, water to binder ratio, chemical composition and porosity. The carbonation rate is also related to the compressive strength of the cementitious product [21]. Higher carbon dioxide uptake for lower compressive strength and higher water to binder ratio leads to higher porosity, which can promote better CO₂ diffusion and achieve higher carbonation efficiency [22–25]. The carbonation rate also increases with the increase in limestone content and the introduction of filler in the cement [26].

External factors are related to the exposure environment: humidity, temperature, pressure and CO₂ concentration [27–30]. Therefore, the natural carbonation process occurs slowly over the years, since carbon dioxide concentration in the atmosphere is about 0.03–0.04% [31].

The cementitious carbonation process has been deeply investigated and the carbonation reactions are well known. However, carbonation rate is still unexplained due to many complex factors [32].

Natural CO₂ exposure environmental conditions are difficult to reproduce, due to it being a very slow process. Exposure conditions are constantly changing, along with the gases present in the atmosphere and their percentages ratio. Thus, accelerated carbonation may not reflect exactly the reality, however a good approximation can be achieved.

Forced and accelerated carbonation are carried out under controlled conditions such as in enclosed chambers and CO₂ diffusion is well distributed on the whole surface of concrete [33]. Different parameters can be applied in lab-scale, such as gas pressure, temperature, relative humidity and CO₂ concentration. However, since many variables
cannot be controlled and assimilated to in situ natural CO$_2$ exposure, especially those related to time, evaluating the carbonation rate and degree of concrete during its service life is complex.

In artificial accelerated forced carbonation, it is possible to control CO$_2$ concentration and increase the carbonation rate of the concrete structures by exposing them to higher CO$_2$ concentrations than the natural concentration in the atmosphere [34,35]. However, the increment of CO$_2$ should not be higher than 20% of CO$_2$ due to pore clogging, which affects CO$_2$ diffusion [36–38].

The carbonation process is also influenced by CO$_2$ concentration and by the Ca(OH)$_2$ and calcium-silicate hydrate (C-S-H) contents, but the carbonation of these compounds does not occur simultaneously [33,39,40]. Portlandite (Ca(OH)$_2$) consists of about 20–25% hydrated Portland cement and it is well known that the expose areas carbonate readily [41]. In the studies of Han et al. [42], calcite content increased when Ca(OH)$_2$ crystals were exposed to CO$_2$. The carbonation occurs by nucleation of CaCO$_3$ on the portlandite surface and this calcite layer formed surrounding the portlandite reduces the in-depth carbonation rate. The carbonation of cementitious materials decreases their total porosity due to CaCO$_3$ precipitation and the formation of a CaCO$_3$ layer covering the surface of Ca(OH)$_2$ may hinder further carbonation of portlandite [41]. On the contrary, C-S-H exposed to CO$_2$ experiences a reduction in volume, since calcium ions are released due to decalcification, which contributes to the development of a larger pore structure in a process of decalcification-polymerization of the C-S-H, forming an amorphous silica gel [43].

Liu and Meng [22] described the carbonation steps of cementitious products. The initial phase of carbonation is the transportation and dissolution of CO$_2$, which releases carbonate ions. Then, Ca$^{2+}$ released in the pores of cement reacts with the cement-hydrated products. This reaction results in calcium carbonate (CaCO$_3$).

Peter et al. [44] indicate that the C-S-H phases start to carbonate as soon as calcium hydroxide stops being available for reaction. At this stage, even though some calcium hydroxide content can still exist, the CO$_2$ gas does not reach it via diffusion; it needs previous solubilization. Another reaction can still occur simultaneously to carbonation: the hydration of concrete. More specifically, C$_2$S (dicalcium silicate) and C$_3$S (tricalcium silicate) react with water (key element for carbonation) to form more calcium hydroxide and C-S-H. Therefore, the reactions occur in this order: first, the carbonation of calcium hydroxide, then, the carbonation of the other compounds, namely C-S-H, and finally the hydration of cement. All these reactions compete for CO$_2$. In terms of consumption, calcium hydroxide consumes the most of CO$_2$, about three times more the amount consumed for C-S-H carbonation.

Therefore, the increment of CaCO$_3$ content is not only due to the carbonation of Ca(OH)$_2$, but also of other cement hydrates (i.e., calcium silicate hydroxide and calcium aluminate hydroxide) that can also carbonate [45], as already mentioned. CaCO$_3$ is formed due the Ca$^{2+}$ and CO$_2$ through the dissolution of calcium hydroxide and the decalcification of C-S-H [5]. Other hydrated cement phases such as ettringite and Ca-Al-Fe monosulfates are also decomposed by carbonation forming CaCO$_3$ [15].

Despite water as a fundamental key in the process of carbonation, there is also water formed during some carbonation reactions, which may have a negative effect. This formed water may block the pores, which prevents CO$_2$ diffusion, and, consequently, decreases the carbonation rate. During the process of natural carbonation three polymorphs of calcium carbonate are formed, namely calcite, aragonite and vaterite. Under natural conditions, carbonation of the cementitious products is a very slow process. Therefore, to investigate the CO$_2$ uptake of the cement-based materials over time, the CO$_2$ concentration in the environment should be increased. In order to simulate the carbonation during its service life it is necessary to accelerate and force the carbonation [20,46]. However, during accelerated forced carbonation only calcite is formed [47].
The following Equations (Equations (1)–(9)) represent the main reactions that occur during carbonation.

\[
\begin{align*}
\text{CaO} + \text{H}_2\text{O} &\rightarrow \text{Ca(OH)}_2 \quad \text{(Calcium hydroxide)} \quad (1) \\
\text{H}_2\text{O} + \text{CO}_2 &\rightarrow \text{H}_2\text{CO}_3 \quad \text{(Carbonic acid)} \quad (2) \\
\text{Ca(OH)}_2 + \text{CO}_2 &\rightarrow \text{CaCO}_3 + \text{H}_2\text{O} \quad \text{(Calcium Carbonate)} \quad (3) \\
\text{H}_2\text{CO}_3 + \text{Ca(OH)}_2 &\rightarrow \text{CaCO}_3 + 2\text{H}_2\text{O} \quad (4) \\
\text{C-S-H} + \text{CO}_2 &\rightarrow \text{CaCO}_3 + \text{SiO}_2 \cdot \alpha\text{H}_2\text{O} \quad (5) \\
\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{CaSO}_4 \cdot \text{H}_2\text{O} + \text{CO}_2 + \text{H}_2\text{O} &\rightarrow \text{CaCO}_3 + \text{CaSO}_4 \cdot \text{H}_2\text{O} + \text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O} + \text{H}_2\text{O} \quad (6) \\
3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{CaSO}_4 \cdot 32\text{H}_2\text{O} + 32\text{H}_2\text{CO}_3 \quad \text{(ettringite)} &\rightarrow 3\text{CaCO}_3 + 2\text{Al}_2(\text{OH})_3 + 3\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + 23\text{H}_2\text{O} \quad (7) \\
3\text{CaO} \cdot \text{SiO}_2 + 3\text{CO}_2 + n\text{H}_2\text{O} &\rightarrow \text{CaCO}_3 + \text{SiO}_2 + n\text{H}_2\text{O} + 3\text{CaCO}_3 \quad (8) \\
2\text{CaO} \cdot \text{SiO}_2 + 2\text{CO}_2 + n\text{H}_2\text{O} &\rightarrow \text{CaCO}_3 + \text{SiO}_2 + n\text{H}_2\text{O} + 3\text{CaCO}_3 \quad (9)
\end{align*}
\]

3. Uptake of CO$_2$ Emissions by Carbonation of Concrete and Other Cementitious Products: Quantification

Cementitious materials are subjected to carbonation during their life cycle. However, the carbonation kinetics are still under investigation [48]. In fact, it is well known that cementitious materials indeed carbonate, but the question is how fast they will carbonate [48]. The carbonation process is related to exposure time and surface. The surfaces of cementitious materials in direct contact with CO$_2$ carbonate promptly, but the carbonation of the interior is slower since the carbonation creates a thin layer that hinders the diffusion of CO$_2$. The transport mechanism of carbon dioxide through the already carbonated layer is difficult and depends on the CO$_2$ content, the type of binder, temperature, pressure and porosity. The carbonation rate slows down with the square root of time [16]. In general, concrete structural buildings are designed to last 50 years without considerable rehabilitation interventions and civil engineering structures are designed to have a lifetime of 100 years. Therefore, it is expected that the demolition of most buildings occurs within 100 years. After concrete is demolished, the carbonation process considerably increases due to the larger uncarbonated exposure surfaces and to the much lower volume of concrete pieces. Therefore, CO$_2$ uptake and the carbonation rate drastically increase [48], following the surface carbonation reactions which are influenced by the thickness of the cement paste and considering that the carbonation rate reduces with the square root of time.

Guo et al. [49] studied the global CO$_2$ uptake by cementitious elements from 1930 to 2019. The authors estimated that 52% of CO$_2$ from the corresponding process fabrication of clinker had been absorbed and sequestered in cementitious products that had been produced between 1930 and 2019. Mortars are considered the most significant contributor of the total uptake, contributing to approximately 58% of the estimated CO$_2$ sequestration.

In order to quantify CO$_2$ uptake the time frame for calculation has to be considered, as well as the use of concrete over time, the end uses of concrete in different types of structures and cement-based products, the amount of concrete/cement used in the different end uses, surface area of the different products and structure details related to the amount of concrete/cement, the exposure environments for the different surfaces and the speed of carbonation (k-value) for different concrete qualities and concrete with different types of cement and in different environments [21]. Table 1 presents the maximum amount of CO$_2$ that different cements can absorb. It was observed that the CO$_2$ uptake capacity is related to the clinker content of the cements. CEM I with 95% of clinker presents the highest amount of carbon dioxide capture.
Table 1. Maximum amount of carbon dioxide that different EN 197-1 cement can absorb, adapted from Sanjuán et al. [21].

| Types            | Type Name                | Notification | Clinker   | CO$_2$$_{\text{max}}$ max CO$_2$/kg Cement |
|------------------|--------------------------|--------------|-----------|---------------------------------------------|
| CEM I            | Portland cement          | CEM I        | 95–100    | 0.49                                        |
|                  | Portland slag cement     | CEM II/A-S   | 80–88     | 0.41–0.48                                   |
|                  |                          | CEM II/B-S   | 65–79     | 0.34–0.41                                   |
|                  | Portland silica fume cement | CEM II/A-D | 90–94     | 0.46–0.48                                   |
|                  |                          | CEM II/A-P   | 80–94     | 0.41–0.48                                   |
|                  | Portland pozzolana cement | CEM II/B-P | 65–79     | 0.34–0.41                                   |
|                  |                          | CEM II/A-Q   | 80–94     | 0.41–0.48                                   |
|                  |                          | CEM II/B-Q   | 65–79     | 0.34–0.41                                   |
|                  |                          | CEM II/A-V   | 80–94     | 0.41–0.48                                   |
|                  |                          | CEM II/B-V   | 65–79     | 0.34–0.41                                   |
| CEM II           | Portland fly ash cement  | CEM II/A-W   | 80–94     | 0.41–0.48                                   |
|                  |                          | CEM II/B-W   | 64–79     | 0.34–0.41                                   |
|                  |                          | CEM II/A-T   | 80–94     | 0.41–0.48                                   |
|                  | Portland burnt shale cement | CEM II/B-T | 65–79     | 0.34–0.41                                   |
|                  |                          | CEM II/A-L   | 80–94     | 0.41–0.48                                   |
|                  | Portland limestone cement | CEM II/B-L  | 65–79     | 0.34–0.41                                   |
|                  | Portland composite cement | CEM II/A-M | 80–88     | 0.41–0.48                                   |
|                  |                          | CEM II/B-M   | 65–79     | 0.34–0.41                                   |
| CEM III          | Blast furnace cement     | CEM III/A    | 35–64     | 0.18–0.33                                   |
|                  |                          | CEM III/B    | 20–34     | 0.10–0.18                                   |
|                  |                          | CEM III/C    | 5–19      | 0.03–0.10                                   |
| CEM IV           | Pozzolanic cement        | CEM IV/A     | 65–89     | 0.34–0.46                                   |
|                  |                          | CEM IV/B     | 45–64     | 0.23–0.33                                   |
| CEM V            | Composite cement         | CEM V/A      | 40–64     | 0.21–0.33                                   |
|                  |                          | CEM V/B      | 20–38     | 0.10–0.20                                   |

Therefore, the amount of Ca(OH)$_2$ in the cement influences the carbonation reactions. Since other types of cement can replace clinkers with additives, the material that can carbonate is lesser than the Ordinary Portland Cement, thus the reabsorption of CO$_2$ is lower. On the other hand, blended concrete carbonates faster than the concrete with only cement, since it presents less Ca(OH)$_2$ available [50]. Lagerblad [48] also relates this increment in the carbonation rate of concrete with cement and supplementary materials to the decrease in content of portlandite and hydration products that can be carbonated. Pade and Guimarães [8] highlighted that the use of other types of cement may affect CO$_2$ uptake, since, for instance, in CEM II, an amount of clinker content is replaced by limestone, which accelerates the carbonation rate. Indeed, the addition of fly ash, slag, metakaolin or silica fume also increases the carbonation rates due to the higher porosity of concrete, which presents greater CO$_2$ penetration, and, consequently, faster carbonation. The replacement of cement by supplementary cementitious materials or fillers reduces the hydration rate and leads to an increase in permeability, which enhances CO$_2$ diffusion at early ages [22,51].

A fixed conservative value was considered in order to estimate the amount of carbon dioxide absorbed during the service life of cementitious materials by Sanjuán et al. [21]. The authors established that 15% or 20% of CO$_2$ emissions during the cement manufacturing is reabsorbed by the cementitious elements during their service life. According to Sanjuán et al. [21], the total amount of carbon dioxide that can be absorbed by concretes and mortars made with Portuguese cements, consumed from 2005 to 2015, was quantified until the year 2100, and the results indicate that the total carbon dioxide uptake is, on average, 14.8% to 19.6% of the total CO$_2$ emissions.
Xi et al. [52] investigated the CO\textsubscript{2} uptake of cement materials during their service life, demolition and secondary use of concrete waste between 1930 and 2013, using an analytical model. The authors estimated the exposed surface area, thickness and time. On average, during their service life, concrete structures can absorb around 16\% of the initial emissions. In short, it is observed that the total CO\textsubscript{2} emissions are 68\% from concrete production and 27\% from mortars. Regarding the CO\textsubscript{2} uptake of all the cementitious products, it is estimated that 43\% of the emitted CO\textsubscript{2} is sequestered and that according to Xi et al. [52], 60\% of this corresponds to CO\textsubscript{2} uptake only by mortars.

Pade and Guimaraes [8] developed a methodology to estimate CO\textsubscript{2} uptake in the life cycle of concrete from the Nordic countries, namely Denmark, Iceland, Norway and Sweden in 2003. The results show that the concrete produced in this year can uptake, during its service life and after demolition, on the whole, over 100 years, around 40\% of the CO\textsubscript{2} emitted due to clinker production.

The authors considered a period of service life for concrete structures of 70 years, and additionally 30 years after demolition. The calculation of CO\textsubscript{2} uptake considered the rate of carbonation and the stability of hydrated products. Additionally, the authors also took into account the strength classes, cement contents and exposed surface area of the concrete structures. The estimated carbonation rate was calculated considering CEM I cement type [8].

Felix and Possan [53] also analysed the balance emissions of CO\textsubscript{2} in concrete structures through cement produced and CO\textsubscript{2} sequestration during the concrete life cycle. The authors stressed that the carbonation of non-reinforced concrete elements is beneficial, since it may compensate for the CO\textsubscript{2} generated in cement production. In conclusion, CO\textsubscript{2} uptake is linked to the type of cement used. Concerning the balance of CO\textsubscript{2} emissions, concrete with blended cement consuming around 400 kg/m\textsuperscript{3} captures approximately 90\% of CO\textsubscript{2} emitted during its production, considering the lifetime and post-demolition.

Possan et al. [27] investigated CO\textsubscript{2} absorption by a concrete dam structure located in Brazil (Itaipu), considered one of the largest engineering projects of the world and built without reinforcement. The authors concluded that considering a period of almost 40 years in service, the concrete structure captured approximately 14,000 tons of CO\textsubscript{2}. This study also verified that the environment and moisture conditions are significant influencing factors, in addition to the strength of the concrete. CO\textsubscript{2} diffusion is very slow at high and low relative humidity [54], since, in saturated conditions, the pores are filled with water and block CO\textsubscript{2} penetration, whereas a dry cementitious matrix does not present enough water for carbonations reactions [55]. It is also important to bear in mind that high temperatures speed up the carbonation of concrete [56].

Jacobsen and Jahren [57] estimate that 16\% of CO\textsubscript{2} emitted due to clinker production is reabsorbed by concrete during its service life of 70 years in Norway. Lee et al. [58] reported a CO\textsubscript{2} uptake of 2.9\% by the cementitious structures over a service life of 20 years. In the studies of Gajda [54,59], it is estimated that an average concrete construction in the United States may absorb approximately 2.1 million metric tons of carbon dioxide in 100 years after construction, which corresponds to 7.6\% of CO\textsubscript{2} emissions in cement production for this country. The authors assumed for each concrete element, defined mix design proportions, thickness and area of exposure surface. These studies concluded that all the concrete from 1950 to 2000s in the United States absorbed about 70 million metric tons of atmospheric CO\textsubscript{2}.

Andrade et al. [60] updated the calculation of carbon storage capacity of Spanish cements. The results showed that cement products in service absorbed 10.8–11.2\% of the calcination emissions, considering the surface exposed area and the volume of the element. The authors verified that the highest CO\textsubscript{2} uptake occurred in unsheltered rainwater conditions and the smallest CO\textsubscript{2} absorption was for indoor conditions.

The CO\textsubscript{2} uptake of a specific building in South Korea was carried out by Yang et al. [61]. The authors calculated that the concrete structure could reabsorb between 5.5\% and 5.7\% of CO\textsubscript{2} during its service life and after being demolished, it could present a CO\textsubscript{2} uptake of
10 to 12%. In short, this CO$_2$ uptake corresponds to 18–21% of the CO$_2$ emissions from the cement Portland production.

AzariJafari et al. [62] estimated the CO$_2$ uptake of concrete in the US pavement network. The authors calculated that up to 5.8 million metric tons (Mt) of CO$_2$ is sequestered by the pavement during its service life and end-of-life. It is important to highlight that all the studies depend on the adopted methodology and assumptions by each researcher, which makes it difficult to compare the results. The results from the literature differ by several factors in concrete carbonation, namely strength, exposure environment conditions, cement type and content, age of the structure, etc. Nonetheless, all the studies show that CO$_2$ uptake by the natural carbonation of cementitious products during their service life exists and partially compensates for the greenhouse gas emissions of cement production. The calculation of CO$_2$ reabsorption also depends on the considered time for a service life of a construction product and if it takes into account the carbonation of concrete element after demolition.

The type of cement element is also an important factor to be considered in order to quantify carbon storage capacity [63]. Table 2 synthesizes the CO$_2$ uptake of buildings’ concrete quantified by previous works; bearing in mind that the calculations refer to different regions and lifetime of the constructions.

| CO$_2$ Cumulative UPTAKE of Building Concretes | Service Life Considered in Years | References |
|-----------------------------------------------|---------------------------------|------------|
| 16%                                           | 40                              | Kim and Chae [64] |
| 19%                                           | 70                              | Jacobsen and Jahren [57] |
| 43%                                           | 90                              | Xi et al. [52] |
| 55%                                           |                                 | Guo et al. [49] |
| 18–21%                                        | 100                             | Yang et al. [61] |
| 24%                                           |                                 | Sanjuán et al. [21] |
| 38%                                           |                                 | Gajda and Miller [54] |
| 40%                                           |                                 | Pade and Guimaraes [8] |

Therefore, most authors estimated that when considering the elements of all concretes during their service life reveals a CO$_2$ uptake between 16 and 32% of the total emissions from cement production. However, regarding cement elements as mortars, the authors verified that this CO$_2$ absorption is much higher than the buildings’ concretes. Xi et al. [52] reported that mortars can absorb an average of 97.9% of the annual initial emissions during their service life. This is due to the high exposure area and thin layers, which contribute to CO$_2$ uptake in short periods, in addition to a higher permeability than concrete. In the study of Sanjuán et al. [21], it is also assumed that a full carbonation in mortars is achieved after 10–15 years of service. Mortars carbonate faster than civil works concrete due to the larger exposed surface area and higher porosity.

From the literature, it can be noted that to estimate CO$_2$ uptake through carbonation of the concrete during its lifetime depends on several factors. However, it is gaining consensus that CO$_2$ absorption of concrete elements should be taken into account in its life cycle, in order to partially offset the impact of its production. The size and exposure conditions of the concrete elements are the most determining factors. Cementitious elements with a larger surface/volume ratio, are more prone to carbonate, i.e., mortars. The external factors such as humidity and temperature may influence carbonation. Overall, it is stated that the concrete elements indicate CO$_2$ uptake potential not only during their service life, but also after demolition.

4. Carbonation of Recycled Aggregates

The use of recycled aggregates (RA) from construction and demolition waste (CDW), as replacement for natural aggregates (NA), has been considered as one of the
most effective approaches for recycling and contributes to greater sustainability [65,66]. Moreover, CDW may have potential to capture and store CO₂ due to its alkalinity, to hydroxides and calcium and magnesium silicates available and the larger exposure surface area [1,2,49,67] in comparison with built structures. In general, the carbonation of concrete is limited to a relatively thin layer near the exposed surface due to the fact that the carbonated layer reduces CO₂ diffusion. Therefore, CO₂ uptake of concrete during service life calculations, previously mentioned, indicate that it still presents high carbonation potential when it is demolished [36,68–70]. The crushed CDW presents a large source of non-carbonated old concrete, which contributes to the increase in carbonate minerals. The CO₂ uptake significantly increases after the demolition of concrete [23]. Furthermore, CO₂ sequestration can improve the quality of CDW [2,5,71], since this rapid carbonation increases the particle density and strength and decreases the water absorption [67,72,73]. Pamenter and Myers [74] highlighted that the CO₂ uptake of the cementitious elements after the end-of-life depends on how the CDW is processed; if these materials are deposited in landfill, their CO₂ absorption is lower than those crushed and recycled.

The particle size of the RA is an influencing factor in the rate of carbonation. The CO₂ uptake in CDW significantly increases for smaller particle sizes due to the higher specific exposure area of CO₂ [4,45,55,75–77]. In the study of Ueno et al. [70] the smaller particles captured more CO₂ during exposure than the coarser ones. Another influencing aspect is the preparation conditions of RA before carbonation: dried or wetted and dried. Kikuchi and Kuroda [45] reported that the CDW carbonated faster when it was alternatively wetted and dried, since the wetting initiates the dissolution of Ca(OH)₂ and drying allows the diffusion of the CO₂ solution, increasing the carbonation rate. Zhan et al. [55] optimized the moisture of CDW before carbonation by soaking it in water for 2 h, thus increasing the carbonation percentage.

Nedunuri et al. [68] analysed the carbonation potential of the recycled concrete fines at different ages. Concrete CDW were collected at ages of 1, 4, 25, 35 and 60 years of the building at the time of demolition. The CDW was collected immediately from the demolished site, and then crushed, stored and protected from the air environment. The results indicate that in all samples, carbonated phases such as calcite, vaterite and aragonite were present. Portlandite was not observed in the samples over 4 years old, which is probably due to the carbonation reactions that occurred before and have consumed all the available Ca(OH)₂. The carbonation potential of CDW was measured through thermal analysis and the results showed that even though the CO₂ capture depends on the age of the concrete when demolished, all the CDW still present carbonation potential. The authors estimated that 16–48% of CO₂ emitted was captured by the concrete during the service life of the structure. It is estimated that the CO₂ uptake increases around 2.4 times when the concrete structure is demolished [8]. Dodoo et al. [56] estimated that nearly 50% of CO₂ emissions are captured by concrete after being crushed and exposed for one year.

Pade and Guimaraes [8] analysed the CO₂ uptake of four different Nordic countries. The authors found out that in the countries where the demolished concrete is crushed, the carbonated rate significantly increases when compared to the countries that place the demolished concrete in a landfill, since the crushing process rapidly increases the surface area of the recycled concrete. The CO₂ uptake increase after demolition is between 30 and 50% during thirty years.

Several studies have quantified the CO₂ uptake of CDW. In the work of Kikuchi and Kuroda [45], it was reported that 1 ton of recycled crushed stone could take up 11 kg of CO₂, while Xuan et al. [4] verified that 1 ton of RA from CDW sequestered about 8 kg of CO₂. Other studies analysed the CO₂ uptake by recycled concrete aggregates [67,72,78,79]. The results indicate that CO₂ uptake was faster for thinner particles.
Mazurana et al. [80] investigated CO₂ uptake by rendering mortars produced with recycled aggregates from CDW. The authors verified that the full replacement of natural sand by RA increased the CO₂ capture up to 57% of the cementitious element. Andrade et al. [81] also studied the CO₂ uptake of mortars with replacement of recycled aggregates. The results showed that for the mortars with RA the CO₂ sequestration increased when increasing the RA content, since RA present higher alkalinity compared to the natural ones and modified mortars are also more porous, which facilitates CO₂ penetration.

In summary, CO₂ sequestration through CDW has been found to be a promising solution to reduce the emission of greenhouse gases and achieve a better CO₂ balance cycle for cement and construction industries. The use of carbon dioxide capture and storage techniques in the cement sector is necessary to maintain CO₂ emissions at a low rate.

5. Conclusions

From this review, the following conclusions can be drawn:

- In terms of the CO₂ capture capacity of the concrete structures in service, it is necessary to take into consideration several influencing factors, including concrete permeability, type of cement, water to binder ratio, CO₂ concentration and time and environment of exposure. Therefore, it can be expected that concretes produced with blended cements have a higher carbonation rate than Portland cement. Moreover, concretes with a higher porosity present greater CO₂ penetration, thus faster carbonation. Relative humidity should be around 50–60% to increase the natural carbonation rate of the concrete. The studies analysed concretes with compressive strengths between 15 and 35 MPa and showed that concrete with a lower compressive strength increased the natural carbonation rate.

- In general, it is reported by several studies that around 19–55% of the CO₂ emitted is captured by the cementitious elements. The type of cementitious element is also an influencing aspect, since rendering mortars present a higher CO₂ uptake than civil works and building concretes, due to the larger surface exposed to the environment and to higher porosity. Even though the mortars consume approximately only 30% of all the cement produced, they sequestrate around 60% of the CO₂ captured by the cementitious materials.

- Regarding the CO₂ capture of CDW aggregates, it has been reported that the carbonation rate of demolished concrete is faster than during service life, since after demolition, the exposed surface drastically increases. Concrete after demolition can increase CO₂ uptake by up to 50% in comparison with the structures in service. Influencing factors such as particle size, cement content and relative humidity affect the carbonation. Thinner recycled aggregates carbonate faster than the others. The age of the building at demolition time is also important, since old constructions are prone to be more carbonated. The time that the recycled aggregate has been stockpiling also increases the carbonation percentage, since the more time the CDW is in contact with the air, the more fully carbonated the particle will be. Generally, considering several studies, recycled concrete aggregate from demolition captures about 10 kg of CO₂ per 1 ton of waste.

Overall, the CO₂ uptake of cementitious structures in service and post-demolition is influenced by several factors. However, the carbonation potential can partially compensate the carbon dioxide emissions of clinker production, released by the calcination process and energy usage attributed to manufacturing. Even though it seems difficult to quantify the amount of CO₂ reabsorbed by cement products, there are some approaches that allow this quantification over time. Cementitious products uptake CO₂ during their service life; however, this uptake considerably increases after demolition; published results indicate an increase of up to 50%. Thus, the potential of CO₂ capture of cemen-
titious elements after demolition should be used by the industry in order to reduce its carbon footprint.

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