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# Accelerated Carbonation Curing as a Means of Reducing Carbon Dioxide Emissions

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## Abstract

Globally, carbon dioxide concentration has immensely increased post the industrial revolution. With more greenhouse gases generated from human activities, more radiation is being absorbed by the Earth's atmosphere, causing an increase in global temperature. The phenomenon is referred to as the greenhouse gas effect. Alone, the cement industry contributes to approximately 5–8% of the global greenhouse gas emissions. Scientists and environmentalists have proposed different scenarios to alleviate such emissions. Among these, accelerated carbonation curing has been advocated as a promising mechanism to permanently sequester carbon dioxide. It has been applied to numerous construction applications, including concrete masonry blocks, concrete paving blocks, ceramic bricks, concrete pipes, and cement-bonded particleboards. Experimental results have shown that not only does it significantly reduce the carbon emissions, it also improves the mechanical and durability properties of carbonated products. The process enhances material performance, offers environmental benefits, and provides an excellent means to recycle carbon dioxide.

**Keywords:** carbonation curing, construction applications, mechanical properties, durability performance, environmental benefits

## 1. Introduction

Greenhouse gases are responsible for maintaining ecological balance and warmth on the planet. Of the total greenhouse gases, carbon dioxide is the main component comprising about 76% [1, 2]. With more CO<sub>2</sub> generated from industries, urbanization, and human activities, more radiation will be absorbed by the Earth's atmosphere, causing an increase in global temperature. The phenomenon is referred to as the greenhouse gas effect. In the 1990s, the rise in the planet's average temperature was 0.74°C. By the end of the 21st century, it is projected to increase by up to 6.4°C [3], instigating cataclysmic changes, as melting of polar ice, increase in sea levels, variations in rainfall and relative humidity (RH), and disappearance of fauna, among others [4].

Of the emitted carbon dioxide gas, the cement industry is responsible for about 5–8% [5]. Such emission is associated with the calcination of limestone (CaCO<sub>3</sub>) to produce lime (CaO) and CO<sub>2</sub> and the burning of fossil fuels for clinkering and grinding. Indeed, it is estimated that the production of one ton of cement releases an equal weight of CO<sub>2</sub> gas [6]. Cement is the main constituent of concrete, the world's most

consumed man-made material with approximately one cubic meter being produced per capita [7]. With the rise in the human population, there is an ever-increasing demand for infrastructure and superstructures. Accordingly, more cement and concrete will be needed. As a result, cement production is becoming an increasing global pressing issue from an ecological, social, and environmental standpoint. To alleviate the emission of CO<sub>2</sub> associated with producing cement and concrete, scientists and environmentalists have proposed different schemes, including the replacement of cement with supplementary cementitious materials (SCMs), increase in energy efficiency, use of alternate fuels, and carbon sequestration [8].

The first scheme proposes modifications to the mixture proportions by replacing cement with SCMs, which are typically industrial waste materials. Cement kiln dust, a by-product of cement manufacturing, has been utilized in producing sustainable composites for construction applications with economic and environmental benefits [9–11]. Other industrial by-products, including fly ash, slag, rice husk ash, limestone filler, and silica fume, have also been used as partial cement replacement in the production of sustainable concrete [12–17]. The properties of so-produced concrete are equivalent, if not superior, to those of conventional cement-based counterparts. Furthermore, efforts have been made to fully replace cement in mortar and concrete. The resulting product has been denoted as alkali-activated or geopolymer mortar/concrete. Numerous studies have investigated the fresh and hardened properties of this novel material and have provided valuable input on its contribution to sustainable construction [18–43]. Nevertheless, the availability and innate compositional variability of the industrial by-products pose a challenge to the adoption and progression of this CO<sub>2</sub> mitigating strategy.

Cement-related carbon emissions could also be reduced by increasing energy efficiency or utilizing alternative fuels during cement production. The use of blended cements, high-activation grinding, and high-efficiency separators, driers, calciners, and clinker cools have been reported to significantly reduce the CO<sub>2</sub> emissions and energy requirements by the cement industry [44–47]. Yet, the suggested modifications in this scheme may not always be practical or economically feasible. Conversely, some studies aimed to alleviate the carbon emissions associated with the generated thermal energy by replacing fossil fuels with alternative fuels, including scrap tires, biomass residues, waste oils, plastics, slaughterhouse residues, spent pot lining, and sewage sludge [48–53]. The scenario is considered environment-friendly, as it conserves natural resources and recycles industrial wastes [54]. However, the different characteristics of these alternative fuels compared to fossil fuels have led to uneven heat distribution, unstable precalciner operation, and dusty kilns, among other complications [55].

While CO<sub>2</sub> emissions could be substantially reduced using the first three methods, they may not always be practical, feasible, or reliable. On the other hand, carbon sequestration or storage has been shown to be a more viable scheme due to its applicability to stationary point sources over the short term. Geological and ocean storage have been mainly practiced for the past few decades [56]. Nevertheless, mineral sequestration has shown great potential, specifically through the accelerated carbonation of hardened cement and concrete.

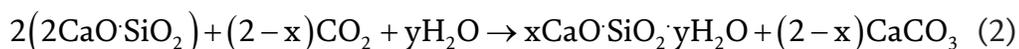
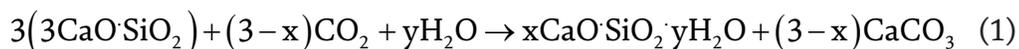
Carbonation is a curing mechanism applied to fresh concrete, i.e. within the first 24 hours after casting. It entails an exothermic chemical reaction between CO<sub>2</sub> and calcium-carrying compounds in cement. Its advantages are three-fold: 1) rapid strength gain, 2) enhanced durability performance, and 3) permanent sequestration of carbon dioxide gas [57, 58]. This chapter summarizes the research and experimental findings of collective studies that have utilized accelerated carbonation in construction applications, including concrete masonry blocks, concrete paving blocks, concrete pipes, reinforced concrete beams, cement-bonded particleboards,

and ceramic bricks. Other topics are also covered, comprising the fundamentals, processes, characterization techniques, and environmental benefits of carbonation of concrete. This work aims to shed light on the technical and environmental gains of accelerated carbonation and its applicability to different construction applications as a means of reducing cement-related carbon dioxide emissions.

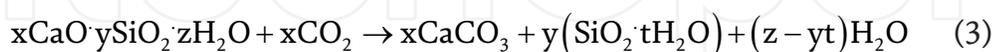
## 2. Fundamentals of carbonation

### 2.1 Reaction kinetics

Carbonation is a physicochemical reaction between cement and carbon dioxide gas in aqueous conditions. More specifically, it is the calcium silicates and their hydration products that undergo carbonation. At early age, calcium silicates, in the form of alite ( $3\text{CaO}\cdot\text{SiO}_2$ , or  $\text{C}_3\text{S}$ ) and belite ( $2\text{CaO}\cdot\text{SiO}_2$ , or  $\text{C}_2\text{S}$ ), react with  $\text{CO}_2$  in the presence of water to produce calcium silicate hydrate ( $x\text{CaO}\cdot\text{SiO}_2\cdot y\text{H}_2\text{O}$ , or C-S-H) and calcium carbonate ( $\text{CaCO}_3$ ). The reaction is primarily dependent on the rate of  $\text{CO}_2$  diffusion, which, in turn, is controlled by the concentration of  $\text{CO}_2$  and its pressure during carbonation [59]. The exothermic reactions are shown in Eq. (1) and (2) [60, 61].



Carbonation is an accelerated hydration reaction; yet, it is different than typical hydration of cement with water, as calcium carbonate formed instead of calcium hydroxide ( $\text{Ca}(\text{OH})_2$ ) [60, 61]. In Eq. (1) and (2), the values of  $x$  and  $y$  depend on the extent of reaction, whereby the theoretical maximum degree of reactivity is 50% of the cement mass [62]. In the event of extensive carbonation exposure,  $\text{CO}_2$  may decalcify the newly-formed C-S-H to produce silica gel ( $\text{SiO}_2$ ) and calcium carbonate, as per Eq. (3) [63]. However, this reaction is not likely to occur in the short-term accelerated carbonation curing regimes employed in the studies addressed herein.



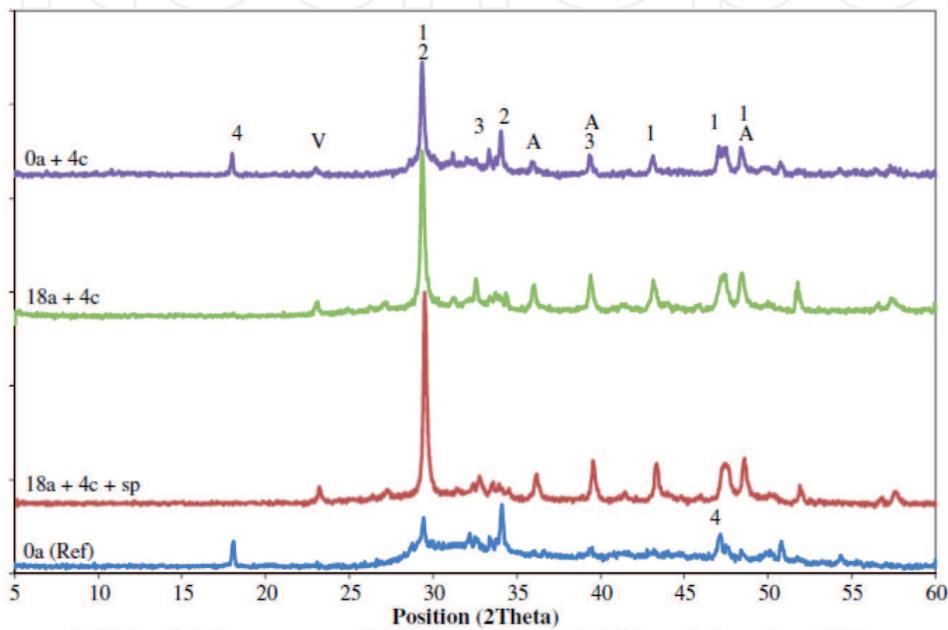
Moreover, the carbonation reaction does not consume all  $\text{C}_3\text{S}$  and  $\text{C}_2\text{S}$  particles, allowing for their subsequent hydration in a post-carbonation moist curing environment [64]. Accordingly, the end-result cementitious matrix is an intermix of  $\text{C}_2\text{S}$ ,  $\text{C}_3\text{S}$ ,  $\text{CaCO}_3$ , C-S-H, and  $\text{Ca}(\text{OH})_2$  [65–70].

### 2.2 Characterization of the reaction products

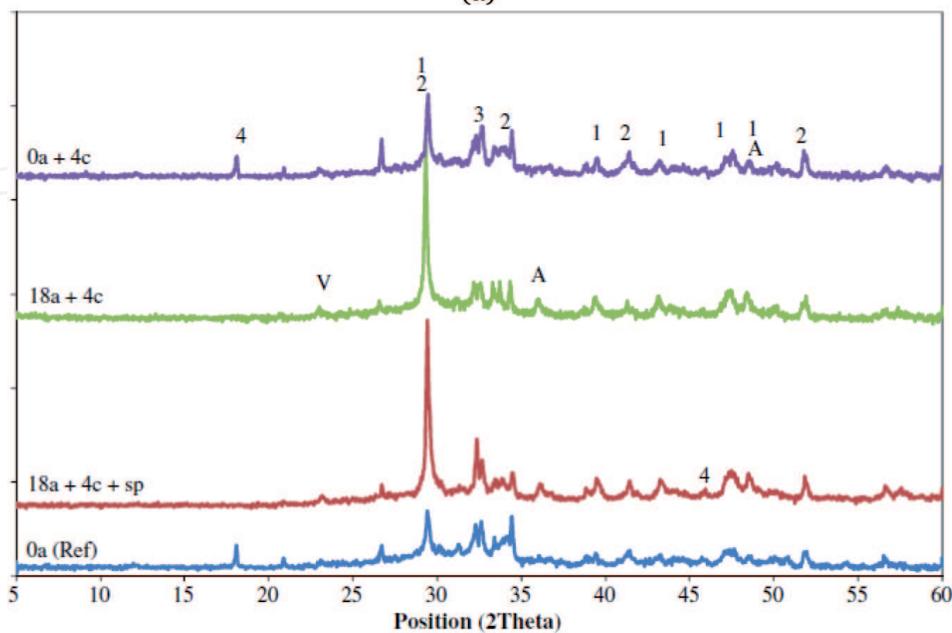
Carbonation of calcium silicates ( $\text{C}_3\text{S}$  and  $\text{C}_2\text{S}$ ) is a form of mineral carbon sequestration that converts carbon dioxide gas into thermodynamically stable calcium carbonate. The calcium carbonate has been detected in three polymorph phases, namely aragonite, vaterite, and calcite [61]. Microstructure analysis showed that the first two polymorphs formed due to carbonation of C-S-H, while the third

one was a product of carbonating calcium silicates [71, 72]. Yet, among the three, calcite has been predominantly identified as the main reaction product of accelerated carbonation. In fact, thermogravimetric analysis (TGA) and X-ray diffraction (XRD) have shown that poorly crystalline aragonite and vaterite transformed into the more stable crystalline calcite polymorph during subsequent hydration [66, 73]. This phenomenon is shown in the XRD spectra of **Figure 1**.

While calcium carbonate has been highlighted as the main carbonation reaction product, C-S-H gel has been identified on fewer occasions. Using XRD, C-S-H was qualitatively detected as a slight increase in the baseline between 25 and 35°2θ [66, 73]. This C-S-H was similar in its amorphous morphology to that formed during typical C<sub>3</sub>S hydration but different in that it was characterized by a lower



(a)



(b)

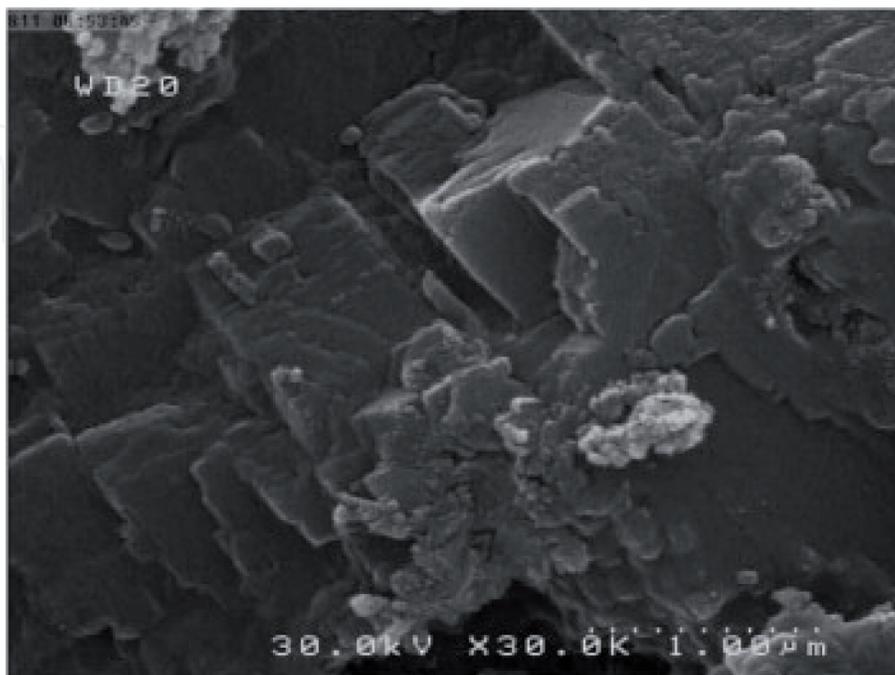
**Figure 1.** XRD pattern of concrete hydration- and carbonation-cured concrete at the age of (a) 1 day and (b) 28 days [66]. Reproduced with permission from the publisher.

CaO-to-SiO<sub>2</sub> ratio [71, 74, 75]. Actually, it was believed that a high degree of carbonation reaction (carbon uptake exceeding 18%, by cement mass) led to intermixing amorphous C-S-H with dominant CaCO<sub>3</sub> to form a calcium silicate hydrocarbonate product, rendering it difficult to be distinctively identifiable [66, 71]. Conversely, lower reactivity (carbon uptake below 10%, by cement mass) integrated small CaCO<sub>3</sub> crystals into a C-S-H-dominant nanostructure [65].

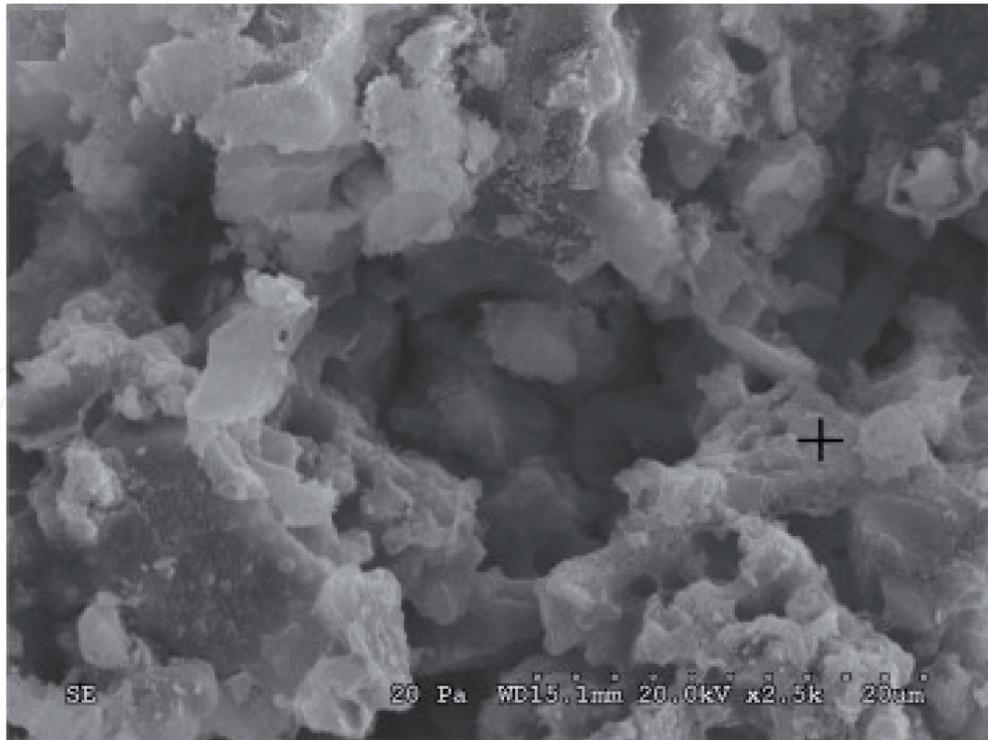
The morphology of these reaction products has also been studied. Calcium carbonate, in its three polymorphs, was reported in different shapes. Cubic, crystal shapes were identified, as depicted in **Figure 2**, when ordinary Portland cement (OPC) concrete was exposed to simultaneous carbonation and chloride ion ingress [76]. Successive preconditioning and carbonation curing of OPC paste and concrete presented amorphous C-S-H gel, calcium hydroxide hexagons, and amorphous calcium carbonate, as shown in **Figure 3a** [65]. A similar morphology is illustrated in **Figure 3b**, whereby OPC concrete made with drinking water treatment sludge was carbonated for 20 hours after 4 hours of preconditioning [77]. Further, an amorphous microstructure with a matrix comprising C-S-H and CaCO<sub>3</sub> was reported when OPC concrete was carbonated for 4 hours after 18 hours of preconditioning (**Figure 4a**). Similar findings have been reported in other studies [68, 78, 79]. Conversely, the morphology of Portland limestone cement (PLC) concrete exposed to a similar carbonation scheme encompassed ball-like forms covered with sharp crystals, as presented in **Figure 4b** [80]. Compared to carbonated OPC concrete, the microstructure of counterparts made with PLC was more porous with higher degree of crystallinity. It was believed that the presence of fine limestone in PLC may have served as nucleation sites for calcium carbonate crystal growth [80].

### 3. Carbonation process

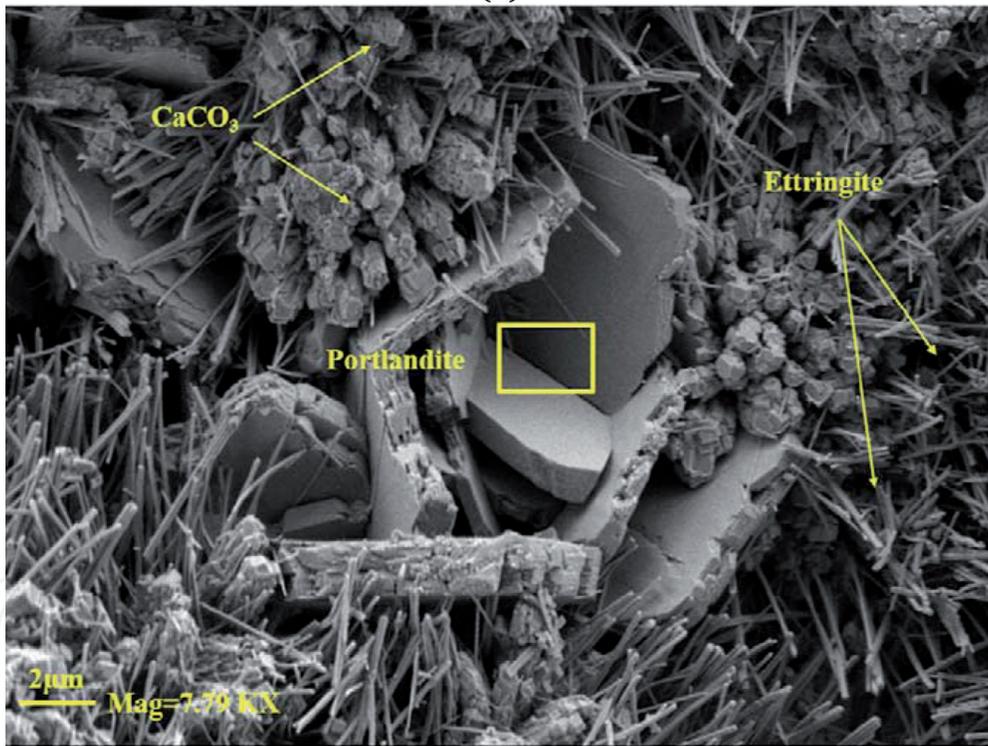
With the advanced understanding of accelerated carbonation, more research has adopted carbonation curing for precast concrete products. The process promises to



**Figure 2.**  
*Morphology of cementitious matrix exposed to simultaneous carbonation and chloride ion ingress [76].  
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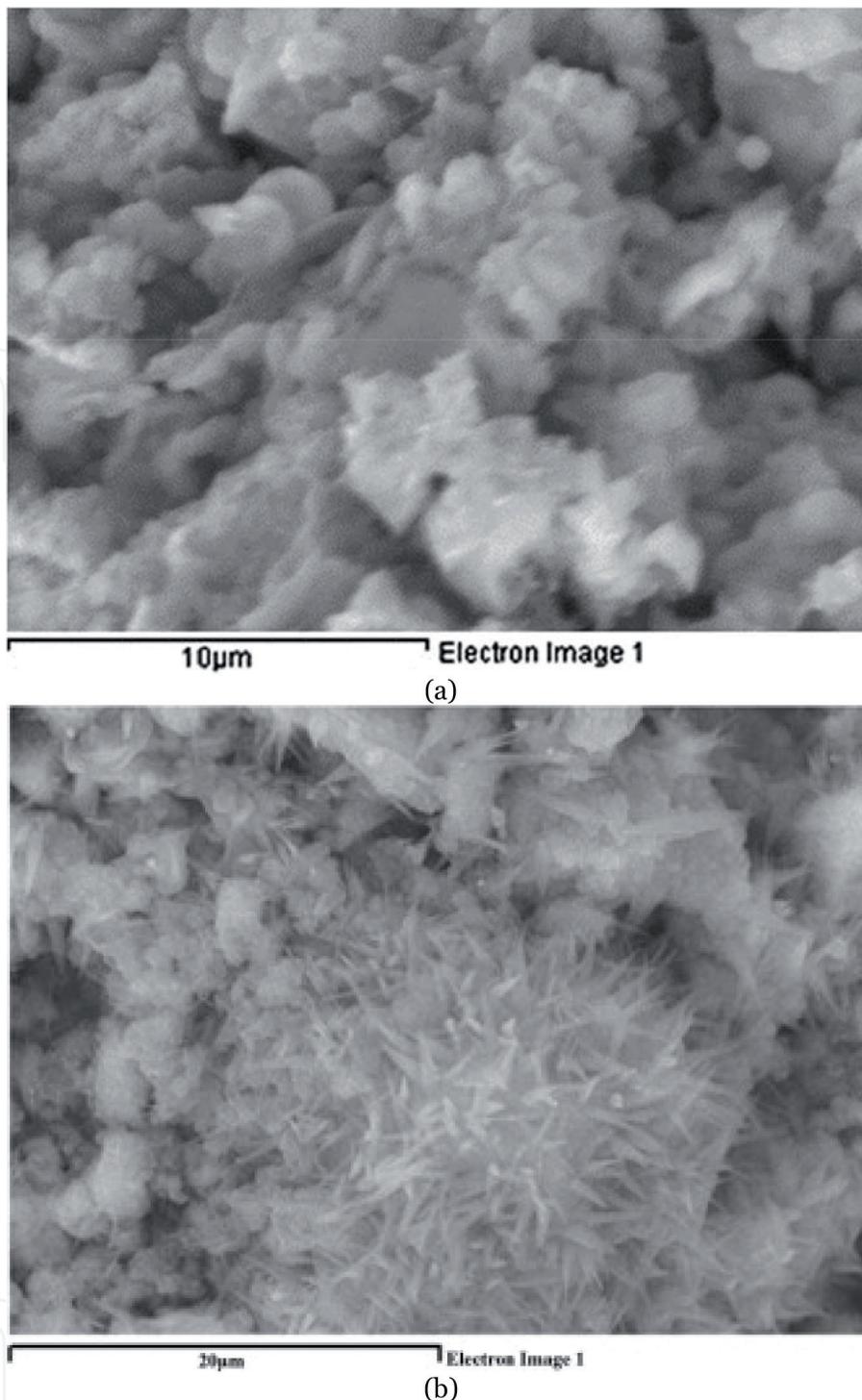
(a)



(b)

**Figure 3.** Morphology of cementitious matrix exposed to (a) 18-hour preconditioning and 2-hour carbonation [18], (b) 4-hour preconditioning and 18-hour carbonation [30]. Reproduced with permission from the publisher.

alleviate anthropogenic emissions through a mineral carbon sequestration technique. However, the environmental impact of carbonation is related to the degree of reaction, which is a function of the availability of water and pore precipitation sites. As such, different curing regimes have been adopted to optimize the amount of water for the highest reaction efficiency. These curing regimes were somewhat different in the adopted duration, temperature, and relative humidity. Yet, they had commonly implemented a three-phase curing process, namely preconditioning, carbonation curing, and post-carbonation hydration.



**Figure 4.**  
SEM micrograph of carbonated (a) OPC concrete [66], (b) PLC concrete [80]. Reproduced with permission from the publisher.

### 3.1 Preconditioning

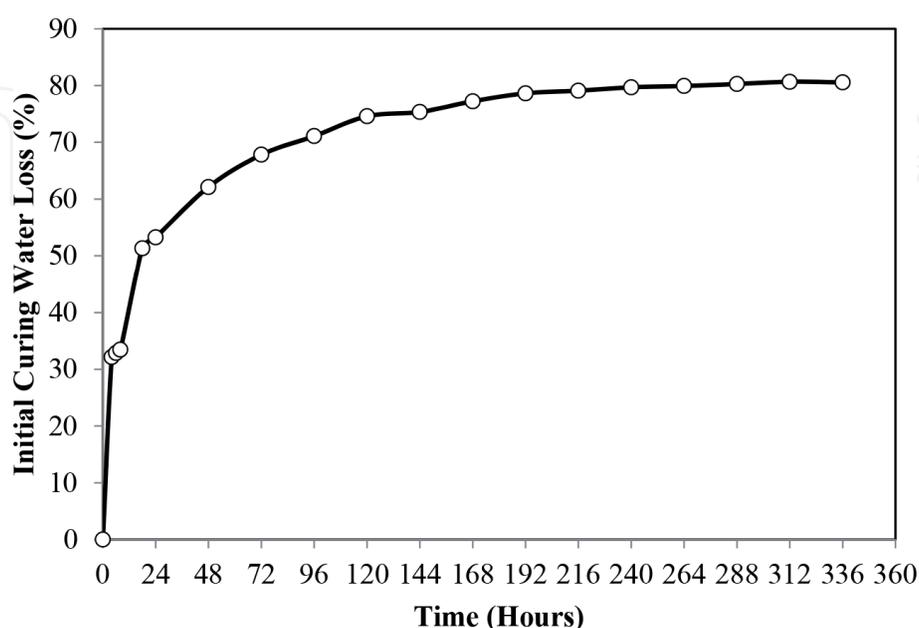
Past studies have reported that free water was necessary to facilitate the dissolution of  $\text{CO}_2$ , however excess water obstructed its penetration through the available porous path [66]. As such, preconditioning was introduced to optimize the amount of water prior to exposing the designated samples to carbon dioxide gas. The adoption of such a process led to a superior carbonation degree and enhanced long-term hydration [81].

For dry mixes, preconditioning took place immediately after casting and before the initial setting of the mix [65, 70, 73, 80, 82]. Conversely, wet mixes were only

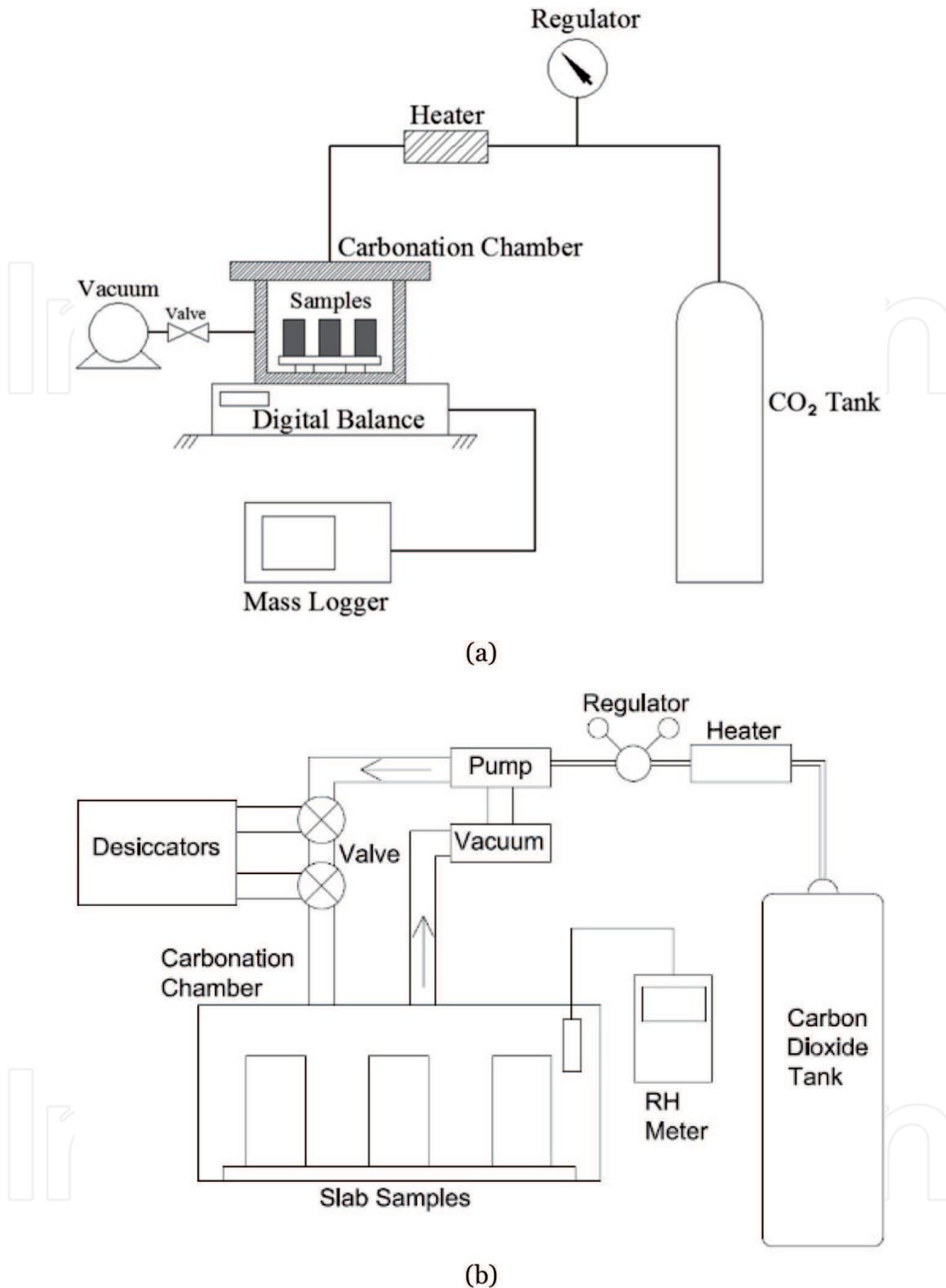
preconditioned after the initial setting [67, 83]. In general, the conditions comprised a duration, temperature, and relative humidity in the ranges of 2–24 hours, 20–25°C, and 40–60%, respectively [66, 67, 70, 73, 80, 82–86]. The effect of preconditioning at 25°C and 50% relative humidity on the water content during 14-day preconditioning is illustrated in **Figure 5**. To simulate industrial practice and limit the total curing time window to 24 hours, a maximum preconditioning duration of 18 hours was recommended [82]. At the end of the preconditioning phase, the cementitious matrix would include anhydrous and hydrated calcium silicate compounds.

### 3.2 Carbonation curing

Carbonation curing encompasses the time period in which concrete is exposed to carbon dioxide gas. Typically, CO<sub>2</sub> is released into a closed chamber and left for a certain duration and under specific conditions for the reaction to take place. A static carbonation system has been typically adopted by most researchers, as shown in **Figure 6a** [66, 70, 80, 82, 85, 87–93]. This carbonation scheme utilized a closed system, whereby the water that evaporated due to the exothermic reaction was included in the estimation of the degree of carbonation. Nevertheless, the reaction was hindered through the precipitation of calcium carbonate particles in the available porous space, leading to a decrease in porosity and retarded diffusivity. To overcome this challenge, a pseudo-dynamic carbonation setup was devised (**Figure 6b**) [73, 94]. This system removed surface free water in a controlled environment and enhanced carbon dioxide penetration by creating a route of capillaries through the sample. It is worth noting that both systems employed a vacuum prior to injecting 99%-pure CO<sub>2</sub> and the pressure was set to 1 bar. Several other researchers used a flue gas to enhance the environmental impact of carbonation, however, the degree of reaction was lesser [67, 95, 96]. Higher pressures of up to 5 bars were also employed [70, 84, 85, 97–99]. Although some promising results were reported when carbonation was utilized at higher pressure, the applicability and feasibility of adopting pressurized carbonation by the industry are yet to be evaluated.



**Figure 5.** Water loss during preconditioning of lightweight concrete [82]. Reproduced with permission from the publisher.



**Figure 6.**  
(a) Static and (b) dynamic carbonation setups [73, 92]. Reproduced with permission from the publisher.

### 3.3 Post-carbonation hydration

The third phase of the carbonation process is the post-carbonation hydration. This step is critical to restore the water lost during preconditioning and the exothermic carbonation reaction and to promote subsequent hydration of unreacted hydraulic cement phases. Early research has reported up to 45% increase in the compressive strength when samples were placed in water for 3 days after

carbonation [64]. Other work incorporated spraying 4-hour carbonated concrete samples every other day until the age of 7 days [82]. The compressive strength increased by 20% compared to carbonated samples left to cure in open air. It is believed that such improvement in mechanical properties is primarily owed to the enhanced pore structure [84].

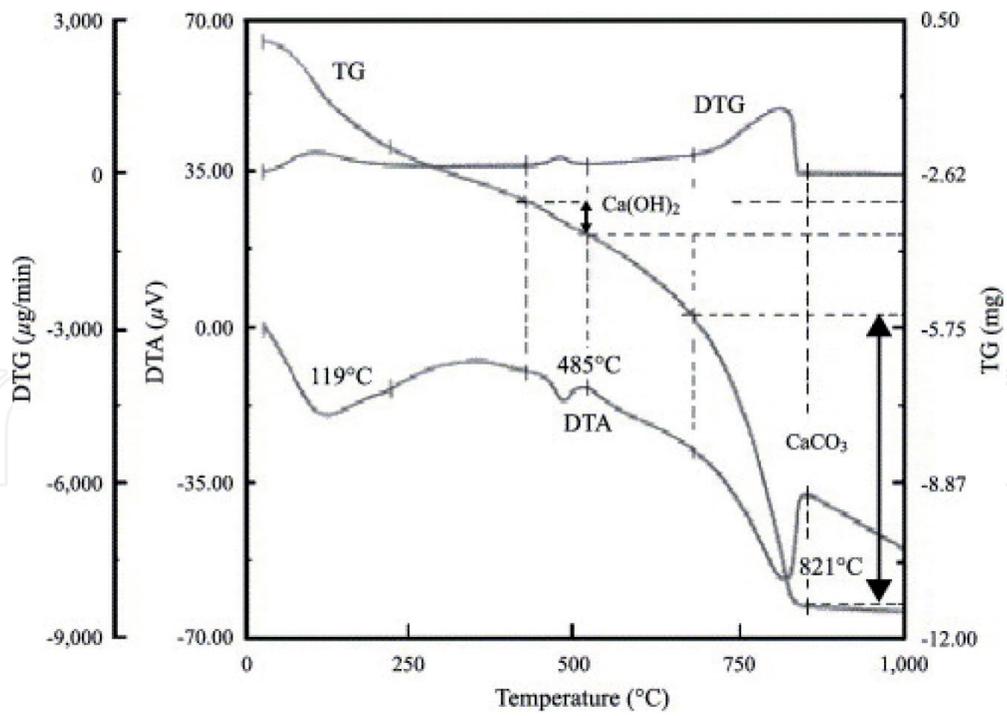
#### 4. Carbonation degree and characterization techniques

Experimental research findings have provided evidence of the feasibility of utilizing carbonation curing for precast concrete products. Yet, the construction industry has not widely adopted it. To promote its utilization and adoption, most past studies aimed to augment the environmental benefit by maximizing the degree of carbonation reaction, which was typically characterized by the carbon uptake. One way to measure the carbon uptake was by examining the mass gained during the carbonation period, assuming homogeneous carbonation across the sample. Because the system was treated as a closed one, the water lost during the exothermic carbonation reaction was collected and added to the final sample mass. Based on Eq. (4), the carbon uptake is the difference in mass between before and after carbonation with the addition of the mass of water lost as a function of the mass of cement. This method has been implemented in several past studies [60, 66, 70, 73, 74, 80, 82, 89, 100–102].

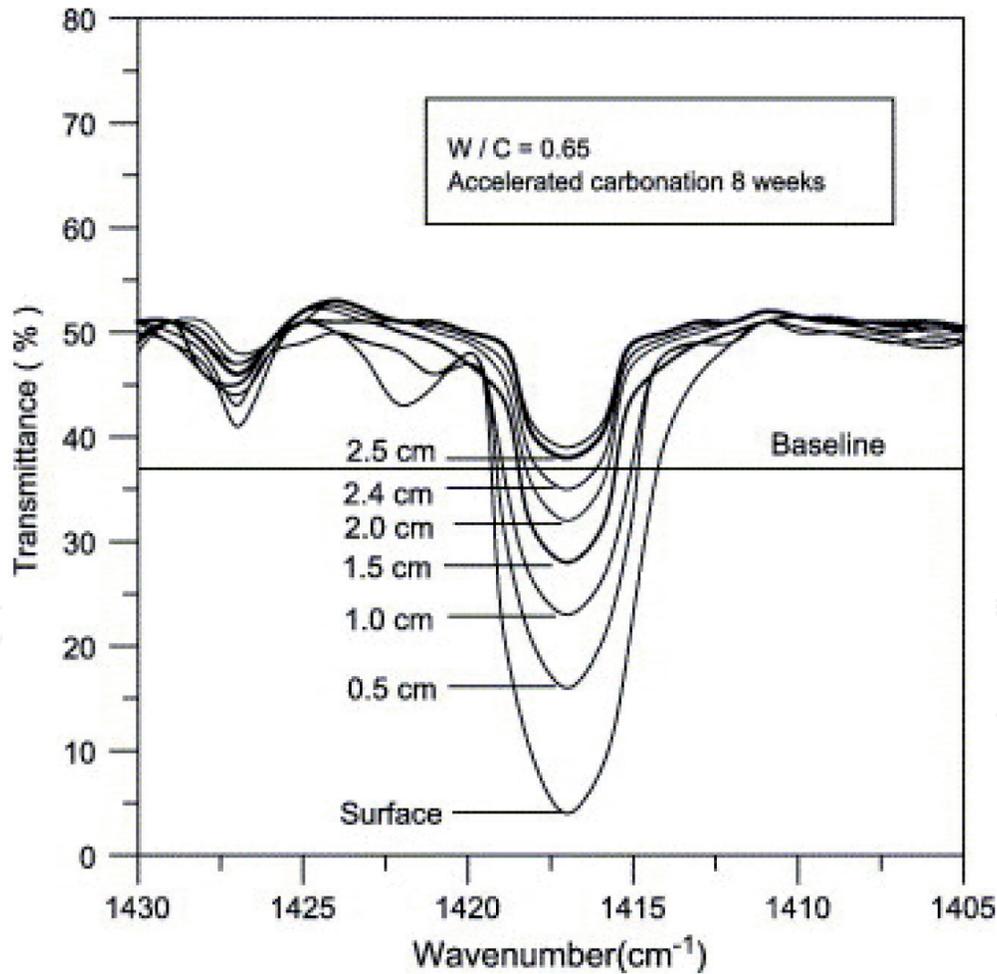
$$\text{Carbon uptake (\%)} = \frac{\text{Final mass} - \text{Initial mass} + \text{water lost}}{\text{Mass of cement}} \times 100\% \quad (4)$$

Thermal analysis is another means of measuring the absolute carbon uptake. In this technique, a thermogravimetric analyzer (TGA) was utilized to monitor the mass loss of a powder sample of carbonated concrete with heat [103]. Alternatively, concrete chunks were decomposed in an electrical muffle furnace by raising the temperature from 25°C to approx. 1000°C. Within this range, multiple hydration and carbonation products were decomposed. The temperature ranges 105–200°C, 200–420°C, 420–550°C, 550–720°C, and 720–950°C were associated with the decomposition of low-temperature C-S-H and ettringite, well-formed C-S-H and C-A-H, calcium hydroxide, poorly crystalline calcium carbonate (vaterite and aragonite), and well crystalline calcite, respectively [66, 71, 73, 80, 81, 104, 105]. However, these ranges slightly differed depending on the type of binder used and the mixture proportions in general, and may even result in an overlap between carbonates and hydrates. To overcome this problem, Fourier transform infrared spectroscopy (FTIR) was employed alongside TGA, as shown in **Figure 7** [106]. It is a vibrational spectroscopic analytical technique that could detect calcium carbonate from the C-O characteristic peak at a wavelength of 1415 cm<sup>-1</sup> [106]. Other analytical tools have also been utilized together with TGA to identify carbonation products, including nuclear magnetic resonance (<sup>29</sup>Si NMR) and XRD [65].

Carbon uptake was also determined by employing coulometric titration in a solution of hydrochloric acid [107]. The technique involved submerging a carbonated concrete powder in the acid solution and measuring the released carbon using the coulometer. The carbon uptake was then obtained using stoichiometric proportions. It is worth noting that thermal analysis and coulometric titration decompose all the carbonates present in the concrete. In an attempt to improve the sustainability of cement, some manufacturers have been replacing



(a)



(b)

**Figure 7.** (a) Thermogravimetric curves and (b) FTIR spectra of carbonated concrete [106]. Reproduced with permission from the publisher.

cement with certain amounts of limestone powder. Such limestone should be deducted from the overall measured carbon content to obtain the absolute carbon uptake [80, 108].

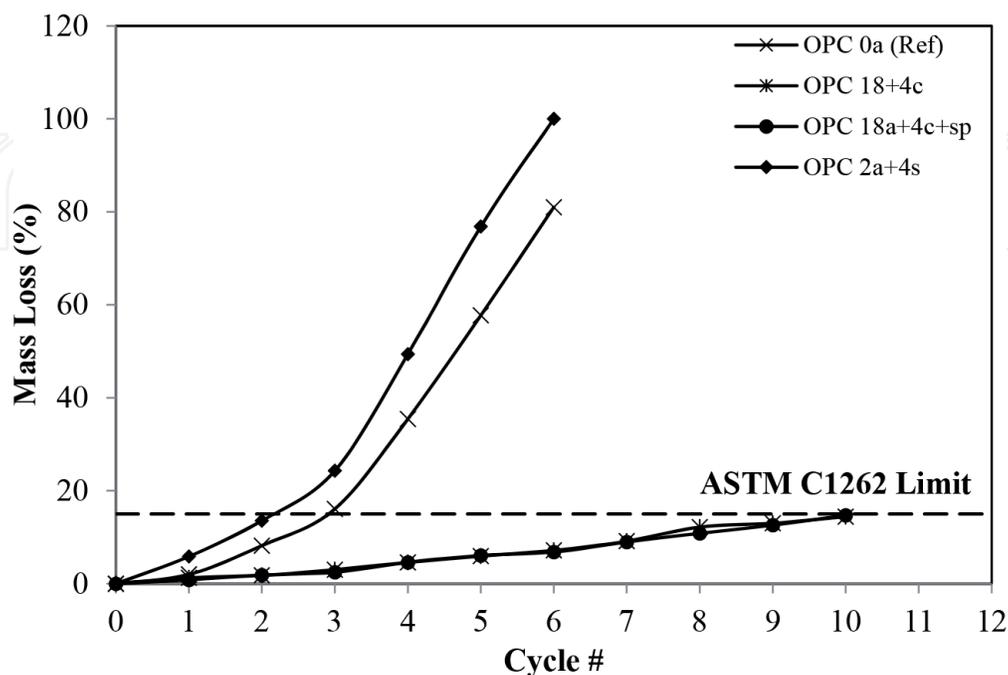
## 5. Carbonation in construction applications

Carbonation curing has been investigated for precast concrete products as a sustainable alternative curing regime to the more typically used steam and moist curing techniques. This section summarizes the collective studies that have examined the effect of accelerated carbonation on the performance of concrete masonry blocks, concrete paving blocks, concrete pipes, reinforced concrete beams, cement-bonded particleboards, and ceramic materials.

### 5.1 Concrete masonry blocks

Accelerated carbonation has been examined as a sustainable curing technique to replace steam curing for concrete blocks. Past studies have examined the mechanical and durability properties of concrete masonry units made with OPC and cured following 0- to 18-hour initial air curing and 2- to 4-hour static carbonation [82, 86, 87, 101, 109, 110]. The carbon uptake reached up to 24%, by cement mass, representing a 48% degree of reaction. The compressive strength within 1 day and at 28 days was comparable to that of steam- and moist-cured counterparts, with values reaching up to 10 and 39 MPa, respectively. It is worth noting that the highest carbon uptake and strength results were noted for samples that were preconditioned for 18 hours in open air prior to carbonation [82, 86, 109, 110]. Additionally, carbonation improved the resistance to chloride penetration by 1.4 and 6.2 times compared to the two conventionally-cured concrete, respectively, and enhanced resistance to sulfate attack by at least 1.5 times [101]. Also, carbonation-cured concrete (18a + 4c and 18a + 4c + sp) had 2 to 3 times better freeze-thaw resistance than concrete cured using steam (2a + 4s) or moist curing (0a), as shown in **Figure 8** [87].

When OPC was replaced by PLC, similar trends related to mechanical properties were noted, but the strength was ultimately lower due to a more porous and crystalline microstructure, as noted in **Figure 4** [66, 80]. Furthermore, concrete blocks



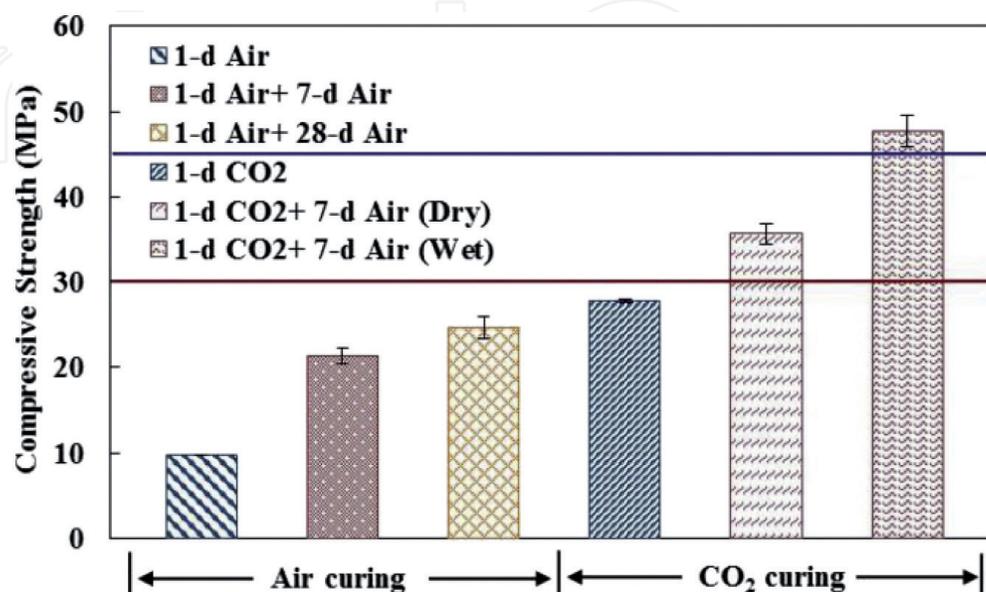
**Figure 8.** Freeze-thaw resistance of carbonated and hydrated concrete [87]. Reproduced with permission from the publisher.

were made with partial replacement of fine aggregates with drinking water treatment sludge and subjected to carbonation [77, 111]. The 1- and 28-day compressive strengths of carbonation-cured concrete blocks were up to 273 and 42% higher than those of normally-cured counterparts, respectively. Splitting tensile strength of the former was also higher than the latter but by no more than 45%. The durability of the concrete blocks was improved, evidenced by the reduction in water capillary absorption and better resistance to sulfate attack. It is believed that this enhancement in durability performance was owed to the pore-filling capacity of newly-formed calcium carbonate [77, 111]. The only detrimental effect of carbonation curing was the increased leaching of aluminum and copper ions, especially for the first 3 days. Nevertheless, the total 60-day leaching concentrations were within the acceptable range, indicating that carbonated concrete blocks made with drinking water treatment sludge were environment-friendly construction materials [111].

## 5.2 Concrete paving blocks

Paving blocks are precast non-reinforced concrete products used in construction applications, including pedestrian and vehicle pavements. With no steel reinforcement and the ability to mass-produce in a precast concrete plant, it is an ideal construction product that could sequester CO<sub>2</sub> through accelerated carbonation curing. In one study, Wang, Yeung [102] examined the use of CO<sub>2</sub> curing to create high performance, low-carbon paving blocks made with contaminated sediment and binary cement. Concrete samples were left to cure in a waterproof membrane until test age, after which they were placed in a drying chamber for 4 hours and cured with CO<sub>2</sub> gas for 24 hours at 0.1 bar above atmospheric pressure. Results of **Figure 9** show that the compressive strength of carbonation-cured concrete blocks was at least 2 times higher than that of air-cured counterparts. Evidently, carbonation curing accelerated the transformation of anhydrous phases into carbonates, while also promoting the formation of more hydrates during subsequent hydration.

Accelerated carbonation has also been employed to cure concrete paver blocks using pure CO<sub>2</sub> and flue gas [112–114]. After preconditioning, concrete samples were exposed to 2 to 4-hour carbonation and then, placed in a mist room to promote subsequent hydration up to 28 days. The CO<sub>2</sub> uptake was reported to be 3.29



**Figure 9.** Compressive strength of concrete paving blocks with various curing methods [102]. Reproduced with permission from the publisher.

and 10.38%, by cement mass, for samples that were carbonated in 20 and 99% CO<sub>2</sub>, respectively. Such lower uptake in the former was due to the lower CO<sub>2</sub> concentration, leading to less CaCO<sub>3</sub> formation and lower compressive strength than the latter. This also resulted in higher water absorption and inferior resistance to efflorescence [112, 113]. In addition, Shao and Lin [114] reported up to 60 times more freeze–thaw resistance when concrete paver blocks were carbonated rather than hydrated.

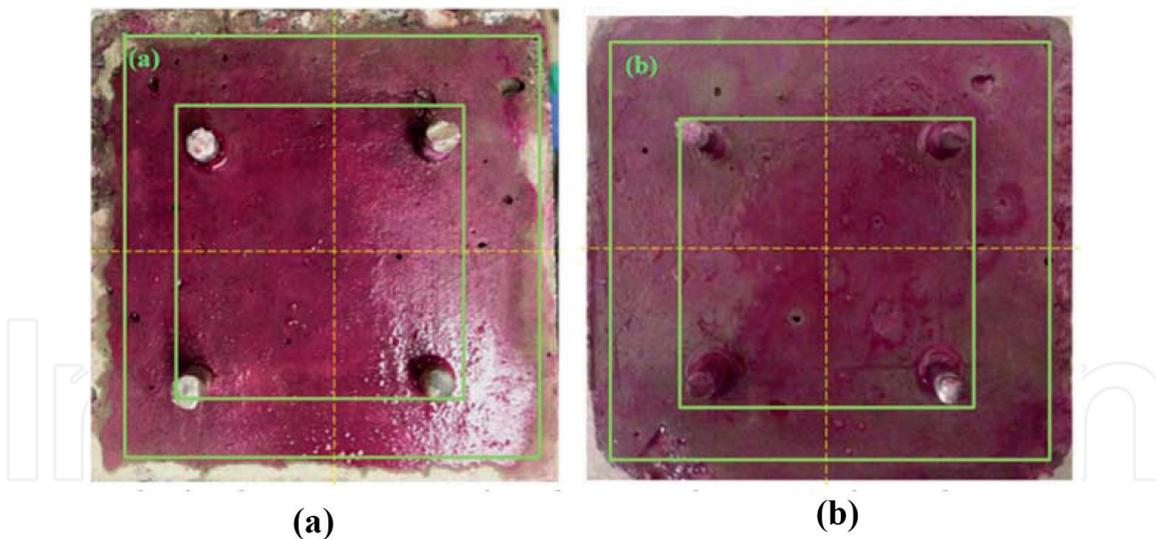
### 5.3 Concrete pipes

Past research has shown that carbonation curing is best applied to fresh concrete directly after casting to promote the chemical reaction between calcium silicates and CO<sub>2</sub> gas. Wet mixes are problematic when demolding within the first few minutes, while dry mixes are ideal for such applications. Concrete pipes are among the different types of concrete products that utilize dry mixes with zero slump. Accordingly, carbonation curing of concrete pipes has been investigated [114]. Samples were cast with a water-cement ratio (w/c) of 0.26 with cement, coarse aggregate, and fine aggregate contents of 426, 853, and 853 kg/m<sup>3</sup>. Directly after casting, they were demolded and placed in a carbonation chamber for 2 hours at a pressure of 1.5 bar and CO<sub>2</sub> purity of 99%. The average carbon uptake was found to be 11.3%, by cement mass. Compared to the hydrated control samples, the carbonated counterparts had a similar 28-day compressive strength of 16 MPa. As concrete pipes may be reinforced with steel, the pH of the carbonated concrete was measured. It was interesting to note that the pH remained above 12, indicating the ability to employ carbonation curing for concrete pipes even in the presence of steel reinforcement.

Other work investigated the feasibility of curing concrete pipes in combined steam and carbonation regime in an attempt to reduce the energy footprint of steam curing, while also sequestering CO<sub>2</sub> [70]. Based on the early-age strength results, concrete pipe samples cured in a combination of the two curing regimes, i.e. steam and carbonation, provided equivalent and superior results to those that were steam and carbonation-cured, respectively, and had a CO<sub>2</sub> uptake of approx. 9%, by cement mass. Compared to samples exposed to steam, those that underwent combined curing showed higher resistance to chloride penetration, sulfate attack, and acid attack, possibly due to the consumption of hydroxyl ions and the formation of calcium carbonate.

### 5.4 Reinforced concrete beams

Despite its adverse effect on reinforced concrete, carbonation of precast reinforced concrete products may be beneficial if performed at an early age. An early-age carbonation curing process was developed for precast reinforced concrete [90, 115]. The detailed curing regime encompassed i) 5-hour in-mold curing at 25°C and 60% RH, ii) 5–6-hour off-mold preconditioning at 25°C and 50 ± 5% RH, iii) 12-hour carbonation curing at a pressure of 5 bars, and iv) 27-day subsequent hydration at 25°C and 95% RH. Carbon sequestration potential was characterized by the CO<sub>2</sub> uptake. It increased from 8 to 15% as the pressure increased from 1 to 5 bar, respectively. This was also associated with an increase in carbonation depth from 8 to 17 mm. Although carbonation decreased the pH of the surface at early age to 9.2, it could recover to 12.3 after 27-day subsequent hydration, evident by the phenolphthalein color profile of **Figure 10**. Evidently, the pH of the area surrounding the steel reinforcement was not affected by carbonation. This indicated that the suggested carbonation curing process posed no risk of corrosion to the



**Figure 10.** Depth of carbonation in a reinforced concrete beam: (a) after carbonation curing; (b) after carbonation curing and 28-day subsequent hydration [92]. Reproduced with permission from the publisher.

steel reinforcement and could be adopted for precast reinforced concrete products. Furthermore, carbonation-cured samples exhibited higher compressive strength, surface resistivity, resistance to chloride penetration, and resistance to weathering carbonation than hydration-cured counterparts. Apparently, carbonation curing reduced the pore size and volume due to calcium carbonate formation and precipitation within the cementitious matrix.

### 5.5 Cement-bonded particleboards

Cement-bonded particleboards are construction products that incorporate cement and fine wood chip fractions. As cement is the main binder in such products, past studies have investigated the use of carbonation curing as a replacement to typical hydration to expedite the production process, while also providing a sink for carbon sequestration. Early studies showed that 2-hour carbonation resulted in a carbon uptake of up to 24%, by cement mass, and compressive strength of 10.5 MPa, which was three times that of the hydrated reference [113, 116]. Nevertheless, prolonging carbonation to 24 hours enhanced the reaction efficiency to obtain an uptake of up to 28% [117]. The resulting flexural strength, freeze-thaw resistance, and wet-dry durability were higher than conventionally-cured counterparts.

Another study employed a wetting-drying-carbonation curing scheme for cellulose fiber-reinforced cement boards [69]. Experimental results showed that accelerated carbonation curing was beneficial to the performance of cement boards. Compared to conventional water curing, it provided superior flexural strength and toughness and reduced capillary porosity and microcracking in the autoclave. Similar findings were reported when cement-bonded particleboards were subjected to supercritical CO<sub>2</sub> curing [118, 119].

In addition to the utilization of carbonation curing to sequester carbon dioxide in cement-bonded particleboards, it has been employed to promote the recyclability of waste materials [120–123]. The mechanical, durability and physical properties of carbonated cement-bonded particleboards were comparable, if not superior, to those of air-cured and hydrated counterparts. Such performance enhancement is owed to the ability of carbonation curing to improve the intactness at the cement-fiber interface, limit the interfacial microcracks, and occupy the capillary space with newly-formed calcium carbonate.

## **5.6 Ceramic bricks**

The applicability of accelerated carbonation curing has been explored in numerous construction applications. The common factor among these applications is the carbonation of calcium or magnesium silicates to produce carbonates. Ceramic materials are rich in such silicates and may be carbonated upon exposure to CO<sub>2</sub>. As such, accelerated carbonation was applied to ceramic bricks from Andalusian factories in Spain [124]. The curing process entailed 24- to 720-hour exposure to CO<sub>2</sub> at a pressure of 10 bars. The authors noted that longer exposure led to higher carbon uptake, with values reaching up to 10%, by ceramic weight. These results highlight the possibility of employing carbonation curing to ceramic waste materials as a means of permanently sequestering carbon dioxide. Yet, more research is needed to validate the findings and evaluate the feasibility of adopting such a technique by the industry.

## **6. Environmental benefit**

Concrete construction applications serve as a potential carbon dioxide sink for CO<sub>2</sub> sequestration. Rather than disposing of CO<sub>2</sub> in geological sites, it can be recycled into concrete with the added benefit of early-age strength and improved durability performance. Concrete products that are typically cured using the steam curing regime can be carbonated to relieve the dependency on high pressure and temperature steam. For instance, a concrete block can sequester nearly 0.5 kg of CO<sub>2</sub>, at an uptake of 24%, by cement mass. At a global annual production of 1800 billion concrete blocks and bricks [125], it will be possible to sequester 900 million tons of CO<sub>2</sub>, which is equivalent to carbon sequestration in approx. 900 geological sites. In comparison, a single concrete paver block could sequester 15.3 g of CO<sub>2</sub>, characterized by an uptake of 10.4%, by cement mass. With 51.4 billion concrete paver blocks (assuming 20% cement content, a thickness of 80 mm, and a density of 2200 kg/m<sup>3</sup>) produced annually [126], these products could sequester up to 1.07 million tons of CO<sub>2</sub>.

Concrete pipes are produced on the scale of 62 million tons per year [127]. At a carbon uptake of 20%, the concrete pipe industry can sequester up to 1.2 million tons of CO<sub>2</sub> per year. Further, precast concrete products in the form of railway ties can store a total of 0.1 million tons of CO<sub>2</sub> per year globally [128]. Conversely, the 9.5 billion m<sup>2</sup> of cement-bonded boards produced annually could sequester 10.8 million tons of CO<sub>2</sub>, assuming 50% cement content, a thickness of 8 mm, a density of 1500 kg/m<sup>3</sup>, and CO<sub>2</sub> uptake of 19%, by cement mass [129]. Although ceramic tiles are different than cementitious concrete, they have also presented a 10% carbon uptake, by total weight. With the global production of 13.6 billion m<sup>2</sup>, and assuming a typical thickness of 1.5 cm, the ceramic tile industry would be capable of sequestering 20.4 million tons of CO<sub>2</sub> [130]. Yet, it should be noted that only one study has been conducted in this research area, signifying the need for further investigation. On a global scale, if all producers of the concrete products presented herein were to adopt carbonation curing, a total of 934 million tons of CO<sub>2</sub> could be sequestered. With an annual global cement production of 4.2 billion tons [131], accelerated carbonation curing could reduce the carbon emissions associated with the cement industry by 22.2%. This capacity could further increase if carbonation were adopted for curing various precast reinforced concrete products.

While the environmental benefit in terms of CO<sub>2</sub> sequestration has been addressed in various research studies, the curing-related water consumption in carbonation curing compared to steam and moist counterparts has not been

investigated yet. Based on the work of El-Hassan, Shao [82], the only water required in accelerated carbonation curing is that for spraying the sample after carbonation. This water promoted subsequent hydration by compensating for the water lost due to preconditioning and the carbonation reaction. As such, the total water consumed in this curing regime was about  $0.085 \text{ m}^3$  per  $\text{m}^3$  of concrete. In contrast, moist and steam curing are estimated to consume about 3 and  $1 \text{ m}^3$  of water for the same volume of concrete, respectively [132]. Clearly, carbonation curing could be deemed more advantageous than moist and steam curing from a water preservation standpoint.

## 7. Conclusions

Accelerated carbonation is an innovative curing regime that promises to expedite strength gain, improve durability performance, and permanently sequester  $\text{CO}_2$  gas in concrete products. Thus, it has the potential to enhance the sustainability of the construction industry.

Reaction kinetics, processes, and final products are comprehensively reviewed. The main chemical reactions occur between calcium silicates ( $\text{C}_3\text{S}$  and  $\text{C}_2\text{S}$ ) in the cement and  $\text{CO}_2$  gas to produce calcium silicate hydrate (C-S-H) gel and calcium carbonate ( $\text{CaCO}_3$ ). Calcium carbonate was detected in its three polymorphic phases, aragonite, vaterite, and calcite, with the former two and latter being associated with the carbonation of C-S-H and calcium silicates, respectively. Their morphology was typical of amorphous, except for the case of carbonating PLC concrete, whereby sharp highly crystalline crystals formed. Conversely, C-S-H was not as easy to detect. In fact, it was intermixed with calcium carbonates to form an amorphous calcium silicate hydrocarbonate product.

The carbonation process was divided into three main stages, preconditioning, carbonation, and subsequent hydration. The utilization of preconditioning was found essential to optimize the water content and promote a higher degree of carbonation reaction. The optimum relative humidity employed in preconditioning was reported as 50–60%. As for carbonation curing, higher reactivity was noted when higher concentration and pressure of  $\text{CO}_2$  were used, evidenced by the higher carbon uptake. Subsequent hydration was introduced afterward to enhance the late age mechanical and durability performance.

The applicability of accelerated carbonation to different construction applications has also been highlighted. Carbonated concrete masonry blocks showed comparable mechanical properties to those of steam- and moist-cured counterparts. Yet, the former's resistance to freeze-thaw damage and sulfate attack was greater than that of the latter. Furthermore, carbonation was applied to concrete paving blocks. The compressive strength and freeze-thaw resistance of carbonated samples were superior to those of hydration- and air-cured equivalents. Similarly, the mechanical and durability performance of concrete pipes and beams subjected to carbonation curing were superior to conventionally-cured counterparts. Also, it was interesting to note that there was no risk of corrosion to the steel reinforcement, as the pH of the surrounding 28-day concrete was above 12. Moreover, the feasibility of employing carbonation as a curing regime for cement-bonded particleboards was assessed. Carbonation curing improved the overall interfacial structure between the cement and fiber and led to the filling of capillary space with newly-formed  $\text{CaCO}_3$ . As a result, enhanced physical, mechanical, and durability properties were reported for carbonated samples compared to conventionally-cured samples. Lastly, carbonation was applied to ceramic bricks as a means of permanently sequestering carbon dioxide.

In addition to its evident improvement in the performance of construction applications, carbonation curing provides a carbon sink to beneficially recycle CO<sub>2</sub>. Yet, its full potential can only be attained if it is adopted on a global scale. The application of carbonation curing to all globally-produced concrete blocks, concrete paving blocks, concrete pipes, cement-bonded particleboards, and ceramic bricks can store up to 934 million tons of CO<sub>2</sub>, leading to a 22.2% reduction in cement-related carbon emissions. Additionally, it has the potential to reduce the water consumed in moist and steam curing by 97 and 91%, respectively. Evidently, the carbonation curing process enhances material performance, offers environmental benefits, and provides an excellent means to recycle carbon dioxide emitted by the cement industry.

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## **Conflict of interest**

The authors declare no conflict of interest.

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