

Journal of Materials and Engineering Structures

Review Paper

A review on carbonation in cement composites and its impacts on durability and potential for CO₂ sequestration

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ARTICLE INFO

Article history :

Received : 22 May 2025

Revised : 2nd October 2025

Accepted : 7 October 2025

Keywords:

Concrete carbonation

Accelerated vs natural carbonation

CO₂ sequestration in concrete

CO₂ uptake

SCMs

ABSTRACT

Carbonation is a key process affecting the durability, strength, and environmental impact of concrete. While it refines pore structure and enhances strength, it also lowers alkalinity, leading to reinforcement corrosion risks. The carbonation rate is influenced by CO₂ concentration, relative humidity, w/c ratio, binder composition, and exposure conditions, all of which determine its impact on concrete performance. This review examines the mechanism of carbonation, influencing factors, effects on mechanical and durability properties, and the relationship between natural and accelerated carbonation. The study highlights that carbonation influences mechanical and durability properties, with research showing that moderate carbonation curing increases compressive strength by 10–30%. Still, excessive carbonation leads to C-S-H decalcification, reducing durability. While accelerated carbonation tests show a carbonation rate 10–15 times faster than natural exposure, the corresponding increase in carbonation depth is only 2.5–5 times, indicating a non-linear relationship that complicates the direct prediction of long-term field performance from laboratory data. Case studies confirm that protected concrete surfaces exhibit minimal carbonation depth (<10 mm in 100 years), whereas exposed surfaces reach up to 50 mm, causing surface deterioration. Additionally, carbonation contributes to CO₂ sequestration, with Itaipu Dam alone absorbing 13,384 tons of CO₂ over 35 years. This comprehensive analysis emphasizes the need for refined, experimentally validated models to accurately predict carbonation and harness its benefits while mitigating risks in future sustainable concrete design.

1 Introduction

Concrete is the most widely used construction material worldwide, valued for its high strength, durability, and cost-effectiveness. However, the production of concrete, particularly the calcination of limestone to produce cement, is a significant source of carbon dioxide (CO₂) emissions, accounting for approximately 8% of global CO₂ emissions annually. As the world grapples with the urgent need to mitigate climate change, the construction industry is under increasing pressure to adopt sustainable practices that reduce its carbon footprint.

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The long-term performance of concrete is significantly influenced by environmental exposure, particularly carbonation, which plays a crucial role in determining both durability and sustainability. [1]. Carbonation occurs when atmospheric CO_2 penetrates the concrete matrix, dissolves in the pore solution, and reacts with calcium hydroxide ($\text{Ca}(\text{OH})_2$) to form calcium carbonate (CaCO_3). While this process densifies the microstructure and enhances compressive strength [2] and surface hardness [3], it also lowers alkalinity, leading to reinforcement depassivation and increasing the risk of corrosion in reinforced concrete structures [4, 5]. The rate and extent of carbonation depend on various parameters, including CO_2 concentration, relative humidity (RH), porosity, water-to-cement (w/c) ratio, and cementitious composition [6–11]. Given the increasing emphasis on sustainable infrastructure, understanding carbonation mechanisms, mitigation strategies, and performance variations under different environmental conditions is critical.

Carbonation affects both the mechanical and durability properties of concrete. In some applications, such as carbonation curing, it is intentionally induced to improve early-age strength and promote CO_2 sequestration [12–14]. However, in reinforced concrete structures, the reduction in pH associated with carbonation accelerates steel corrosion [15], particularly in high- CO_2 urban environments or regions with fluctuating humidity conditions. The microstructural changes caused by carbonation alter pore connectivity [16], influencing long-term performance. Despite extensive research, several uncertainties remain regarding the real-world implications of carbonation under varying exposure conditions, necessitating further investigations.

Although carbonation in concrete has been widely studied, several research gaps and challenges persist. One major challenge is the discrepancy between natural and accelerated carbonation. Laboratory-based accelerated carbonation tests, typically conducted at CO_2 concentrations between 1–10% [17], may not accurately replicate natural carbonation, where atmospheric CO_2 levels are approximately 0.04% [18]. This variation raises concerns about the correlation between accelerated exposure conditions and long-term field performance. Another critical issue is the impact of supplementary cementitious materials (SCMs) on carbonation resistance. While fly ash, silica fume, and ground granulated blast furnace slag (GGBS) enhance durability [19, 20] and reduce environmental impact, they also modify calcium hydroxide availability and porosity, making their effect on carbonation complex and unpredictable. The role of RH in carbonation is also significant, as studies indicate that carbonation occurs most rapidly at RH levels between 50–70% [21–23]. However, its interaction with pore size distribution and permeability in blended cement systems remains poorly understood. Additionally, while early-age carbonation curing improves strength and CO_2 sequestration, its long-term effects on durability, shrinkage, and reinforcement protection require further exploration. Most research on carbonation focuses on controlled laboratory settings, while field studies assessing real-world performance are limited. There is also a lack of comprehensive strategies for maintaining carbonation over time, emphasizing the need for long-term monitoring and effective maintenance techniques.

Despite this extensive research, a critical gap remains in the systematic correlation of findings from accelerated laboratory tests with long-term performance under natural, variable field conditions. This gap is particularly pronounced for modern concrete mixes incorporating high volumes of SCMs, where the interplay between carbonation, changing microstructure, and durability is not fully quantified. Furthermore, there is a need to synthesize the dual role of carbonation as both a durability risk and a carbon sequestration opportunity into practical guidelines for sustainable concrete design.

To bridge these gaps, this review comprehensively analyzes laboratory-based carbonation models with real-world structural performance. It systematically examines the fundamental mechanisms and the influence of critical parameters like mix design and environmental exposure. A key focus is placed on critically evaluating the relationship between accelerated and natural carbonation to improve the reliability of service-life predictions. Finally, the review explores innovative mitigation and utilization strategies, providing a balanced perspective on achieving durable, carbon-conscious concrete infrastructure.

2 Carbonation Mechanism and Microstructural Changes

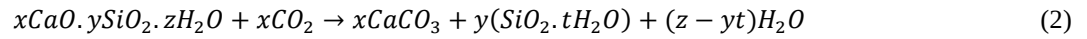
2.1 Overview of Carbonation in Cementitious Materials

Carbonation in cement-based materials is a diffusion-controlled process in which CO_2 penetrates the concrete matrix, initially reacting with the outer surface before progressively moving inward [24]. During this process, ionized CO_2 dissolves calcium ions from hydrated cement phases, leading to the precipitation of CaCO_3 in the pore spaces, which results in a denser microstructure [25]. This reaction is highly exothermic and plays a crucial role in altering the mechanical and durability

properties of concrete. Equation 1 shows the primary reaction, which involves calcium hydroxide (Ca(OH)_2) and CO_2 , forming calcium carbonate (CaCO_3) [26, 27]



In addition, calcium silicate hydrates (C-S-H) react with CO_2 , leading to the formation of CaCO_3 and silica gel [28–30] as per equation 2



However, the exact stoichiometry of C-S-H carbonation remains uncertain, particularly regarding the amount of bound water in silica gel and the polymorphic form of CaCO_3 (calcite, vaterite, or aragonite) that precipitates [26]. This uncertainty makes it challenging to measure porosity evolution due to carbonation accurately.

2.2 Influence of Cement Phases on Carbonation

The primary mineral components in cement clinker include tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A), and tetracalcium aluminoferrite (C_4AF). The carbonation reactivity of these phases follows the sequence: $\text{C}_3\text{S} > \text{C}_2\text{S} > \text{C}_3\text{A} > \text{C}_4\text{AF}$ [31]. Since C_3S and C_2S are the most abundant cement phases, their carbonation dominates, resulting in CaCO_3 and silica gel formation, which occurs faster than their hydration reactions [32]. In contrast, C_3A exhibits minimal carbonation reactivity due to its low affinity for CO_2 [12]. The AFt (ettringite) and AFm (monosulfate) phases, produced by C_3A and C_4AF hydration, decompose under carbonation, forming water, gypsum, alumina gel, and amorphous phases [33], [34]. Notably, C_4AF does not carbonate significantly [35].

2.3 Carbonation Phases: Gaseous, Liquid, and Solid

Carbonation occurs through three distinct phases: gaseous, liquid, and solid [8, 36]. As given in Fig.1, CO_2 diffuses through the air in the gaseous phase and penetrates the concrete surface. In the liquid phase, dissolved CO_2 reacts with water to form carbonic acid (H_2CO_3), dissociating into H^+ and HCO_3^- ions, initiating the breakdown of C-S-H, C_3S , and C_2S . This reaction rapidly drops the pH from 11 to 8, significantly altering the cement matrix. In the solid phase, nucleation and crystallization of CaCO_3 polymorphs occur, producing amorphous calcium carbonate and calcite as the final products [37]. The carbonation front initiates at the exposed surface, where CO_2 concentration is highest, and gradually diminishes towards the core due to diffusion limitations.

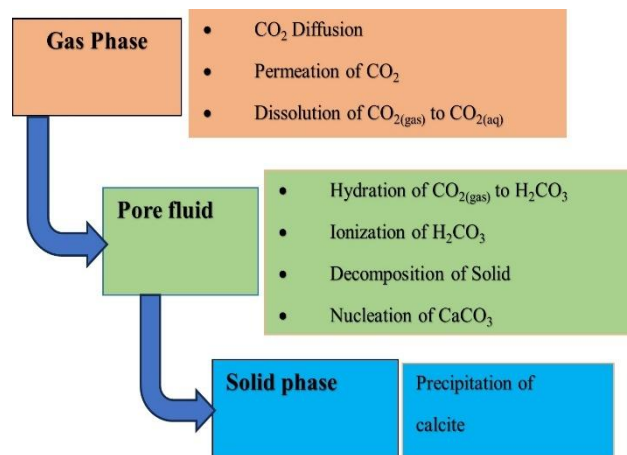


Fig. 1 – Different Phases of Carbonation

2.4 Microstructural Changes Due to Carbonation

The difference in molar solid volumes between hydration and carbonation products leads to microstructural transformations in concrete. The precipitation of CaCO_3 increases solid volume by 11–14%, reducing porosity and refining the pore structure [26, 38]. However, C-S-H carbonation results in calcium leaching, leading to the formation of silica gel, which may cause a local decrease in solid volume and pore refinement [39–41].

When supplementary cementitious materials (SCMs) are added to cement, coarsening of the pore volume has been reported because of the higher amount of C-S-H and the lower amount of CH [42, 43]. However, for both OPC and cement-containing SCMs, some studies have seen a drop in pore volume and refinement of the pore structure [44, 45], whereas other research on the carbonation of Portland cement has revealed a decrease in porosity and an increase in critical pore width diameter [46–48].

2.5 Experimental Characterization of Carbonation

Scanning Electron Microscopy (SEM) and Back-Scattered Electron Imaging (BSE) provide insights into carbonation-induced microstructural changes. The atomic number contrast in SEM imaging distinguishes carbonated and non-carbonated regions, where anhydrous cement grains appear brightest, C-S-H and $\text{Ca}(\text{OH})_2$ appear in light gray, and pores appear dark [49–51]. The microscopic images of OPC samples under various exposure scenarios are displayed in Fig. 2, in which carbonated samples exhibit brighter spots, corresponding to CaCO_3 deposits and decalcified silica gel [51].

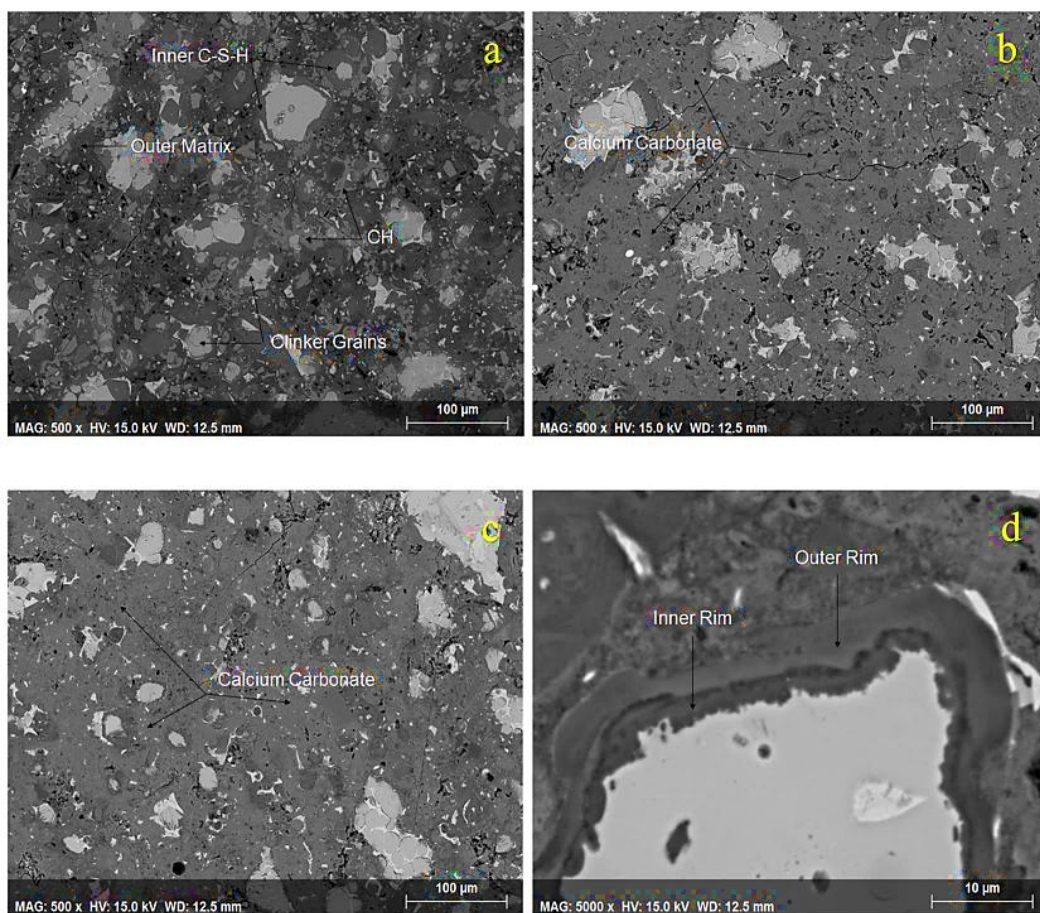


Fig. 2 – SEM images of OPC (a): Uncarbonated, (b) Natural carbonated, (c & d) Accelerated carbonated [51]

Energy Dispersive Spectroscopy (EDS) analysis also confirms Si/Ca and Al/Ca ratio variations, revealing a two-rim structure: inner rim and outer rim (Fig. 2d). Points obtained from the outer rim exhibit lower Si/Ca, while the darker inner rim, i.e., the rim closest to the cement grain, has higher Si/Ca. Elevated Si/Ca and Al/Ca indicate that C-S-H is decalcified and transforms into silica and alumina gel. The outer rim, which appears brighter, corresponds to the inner hydrate before the samples are exposed to carbon dioxide. In contrast, the darker rim is caused by the extra hydration during the exposure period. The formation of calcite, vaterite, and aragonite in carbonated pastes significantly impacts pore connectivity and mechanical properties. Fig. 2 illustrates these transformations, with distinct morphological differences between uncarbonated, naturally carbonated, and accelerated carbonated samples.

2.6 Accelerated Carbonation Techniques

Since natural carbonation progresses slowly under ambient CO₂ levels (~0.04%), accelerated carbonation techniques enhance the reaction rate. Accelerated carbonation chambers control CO₂ concentration, temperature, and RH to simulate long-term carbonation effects within a shorter period. A schematic of a typical accelerated carbonation chamber is presented in Fig. 3.

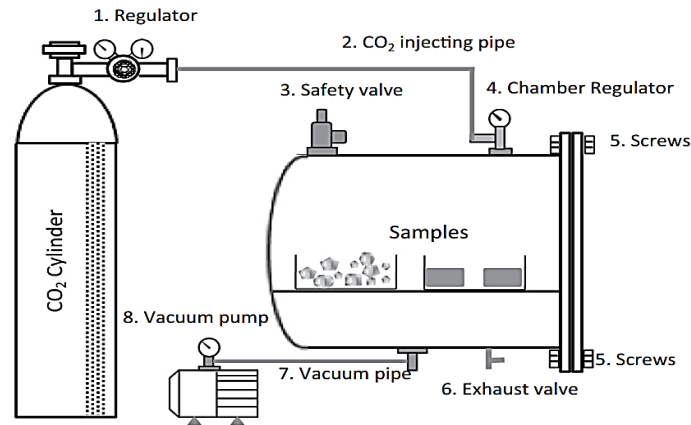


Fig. 3 – Schematic of the accelerated carbonation chamber [52]

For industrial-scale applications, direct utilization of flue gases from cement and steel plants is being explored, offering a cost-effective and energy-efficient approach to carbonation curing. Flue gases contain CO₂ concentrations of 14–33% (cement plants) and 20–30% (steel plants), which can be leveraged for bulk carbonation processes, enhancing both sustainability and material performance [53–55].

3 Carbonation and Its Factors

3.1 Overview of CO₂ Uptake in Cementitious Materials

CO₂ gas is naturally absorbed by exposed concrete, and research done in northern Europe and North America has documented the CO₂ absorbed in concrete through carbonation [38, 56]. Equation 3 is typically used for the calculation of CO₂ absorption [56].

$$CO_2 \text{ absorption (kg CO}_2\text{/m}^3 \text{ concrete)} = 0.75 \times C \times CaO \times M_{CO_2}/M_{CaO} \text{ (kg/m}^3\text{)} \quad (3)$$

Where M represents the molecular weight of the component, and C represents the cement quantity in 1 m³ of concrete (kg/m³).

Lee et al. used Equation 4 to calculate absorbed CO₂ in concrete [57].

$$mCO_2 = [CO_2] \times (A \times B) \times Depth \times 44 \quad (4)$$

where [CO₂] is the concentration of materials in mol/m³ that can react with CO₂ in the concrete, (A × B) is the surface area of the concrete available for air contact, and Depth represents the carbonation depth of the concrete. The molecular weight of CO₂ is 44 g/mol.

3.2 Factors influencing carbonation

3.2.1 Influence of w/c Ratio

Higher water-to-cement (w/c) ratios increase porosity, facilitating CO₂ diffusion and enhancing carbonation depth. Tang et al. [58] observed that samples with w/c = 0.3, 0.45, and 0.55 initially exhibited CO₂ uptake values of 11.9%, 31.3%, and

37.1%, respectively. However, after 90 days of conventional curing, CO₂ uptake reduced to 5.4%, 22.8%, and 18.5%, indicating reduced carbonation at later hydration stages due to pore refinement [59, 60].

Studies indicate that lower w/c ratios result in denser microstructures [61], which limits carbonation depth over time. However, the increase in CO₂ uptake at higher w/c ratios is not entirely beneficial, as excess porosity may also lead to greater permeability, allowing external aggressive agents (e.g., chlorides and sulfates) to penetrate the concrete matrix, reducing durability. While carbonation depth increases with w/c ratio, excessive values (>0.5) decrease carbonation efficiency due to lower Ca(OH)₂ availability [61]. Fig. 4 shows the variation of carbonation depth with w/c ratio.

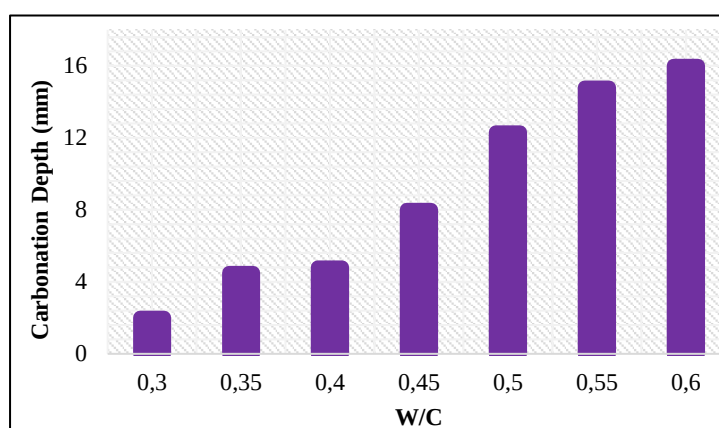


Fig. 4 – Effect of w/c ratio on Carbonation depth [62]

3.2.2 Effect of Curing Time

Jang et al. [63] reported that carbonate formation in alite-rich pastes reached 77% at 28 days, while belite-rich pastes continued to carbonate at a steady rate, achieving 92% carbonation at 28 days. Studies indicate that 50% of total CO₂ uptake occurs in the first 12 hours of exposure, with subsequent decreases due to pore filling and reaction saturation [58, 64]. Zhan et al. found that increasing CO₂ curing time from 0.5 hours to 4 hours led to a higher carbonation percentage of 17.65%, 20.72%, 25.06%, 28.05%, and 28.47%, respectively [65].

Longer curing times improve early carbonation efficiency, but beyond a specific duration, the rate of CO₂ uptake slows due to saturation effects. The findings suggest that most CO₂ sequestration occurs within the first 24 hours, with diminishing returns in prolonged curing conditions. Fig. 5 displays the outcomes of the carbon dioxide uptake by powdered grout at a pressure of 600 psi. The grout initially absorbs carbon dioxide rapidly, growing in mass by more than 8% in the first minute of the reaction and over 15% after many hours. The reaction rate then gradually drops after that. After the first four days of reaction, there was no discernible pickup [66].

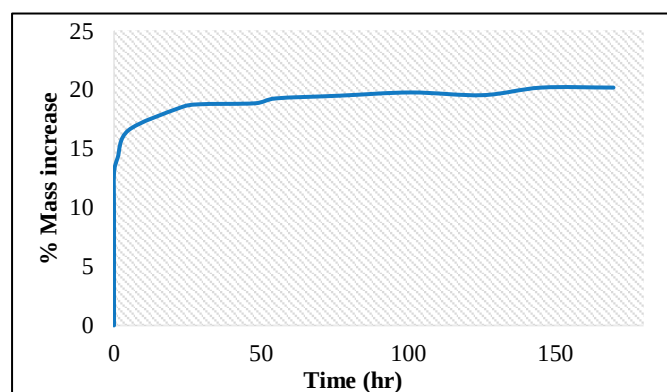


Fig. 5 – Rate of CO₂ uptake vs time [66]

CO₂ curing at early ages can accelerate initial strength gain but impacts long-term carbonation. Samples exposed to 0.5 bar CO₂ pressure for 48 hours exhibited higher early carbonation depth, but as hydration products densify the microstructure, subsequent CO₂ uptake declines. This effect is more pronounced in high w/c ratio mixes, where increased porosity enhances early carbonation but stabilizes over time [65].

3.2.3 Effect of CO₂ Pressure and Concentration

Higher CO₂ pressures enhance carbonation by increasing diffusion rates and C-S-H decalcification. Bishnoi et al. [67] observed that concrete with w/c ratios of 0.35 and 0.45 exposed to 10% CO₂ showed significantly higher carbonation depths than at lower pressures. For several solid grout samples, the impact of CO₂ pressure on the carbonation rate are presented in Fig. 6.

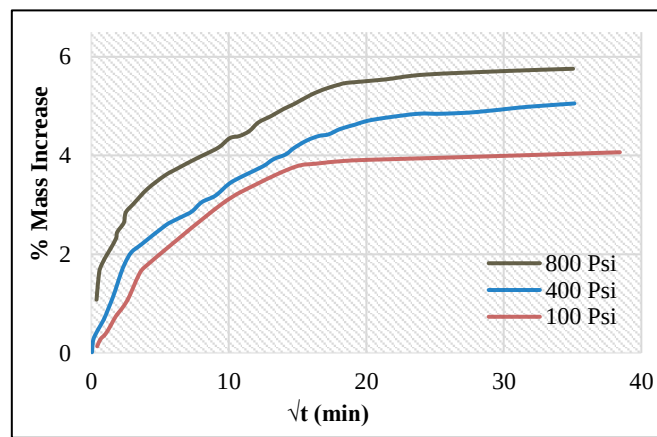


Fig. 6 – The effect of CO₂ pressure on the carbonation rate of solid grout samples [66].

Pu et al. [68] reported that increasing CO₂ from 20% to 40% accelerated carbonation of RCA, but beyond 40%, further increase had minimal effect. Experimental studies confirm that concrete samples exposed to low CO₂ concentrations of 2–20 % for the same carbonation age show a sharp rise in carbonation depth whereas samples exposed to CO₂ concentrations of 20-50 % show a lower increase in carbonation depth. Beyond 50 % of the CO₂ concentration, no subsequent rise in carbonation depth is observed. This is primarily because the carbonation alters the microstructure, making the surface layer denser and reducing the rate of CO₂ diffusion into the concrete [69].

Moreover, increasing the partial pressure of CO₂ from 0.003 MPa to 0.01 MPa leads to significant increases in carbonation rate of mortars, whereas beyond 0.05 MPa, no substantial acceleration is observed [70]. This suggests that an optimal CO₂ concentration exists between 10% and 30% for accelerated carbonation testing without excessive pore blockage.

3.2.4 Effect of Relative Humidity

It has been observed that generally, the carbonation increases as the relative humidity increases, but after a particular value of the RH, the carbonation efficiency is adversely affected. Fig. 7(a) and 8(b) show the effect of RH on the carbonation efficiency. It can be seen that the carbonation efficiency is at its maximum at an RH of 40-70%. At 70% relative humidity, the carbonation depth reaches its maximum value. Equation 5 represents the cubic relationship between RH and the concrete carbonation depth.

$$D = ax^3 + bx^2 + cx + d \quad (5)$$

where D is the carbonation depth of concrete, x is the relative humidity, and a , b , c , and d , are constants [71]. Vagelis et al. observed that the carbonation depth is maximum at a relative humidity of around 50% [72]. When the relative humidity is between 60% and 80%, there is a significant variation in the carbonation depth. Maximum carbonation depth is reached when the relative humidity is between 70 and 75% [73]. The degree of pore saturation likewise drops with comparatively lower RH levels due to the pores holding less water. As a result, rather than the diffusion of CO₂, the chemical reactions determine the rate of reaction. Liquid water can coagulate in micropores when the relative humidity exceeds the optimum level, reducing the quantity of CO₂ transmission channels.

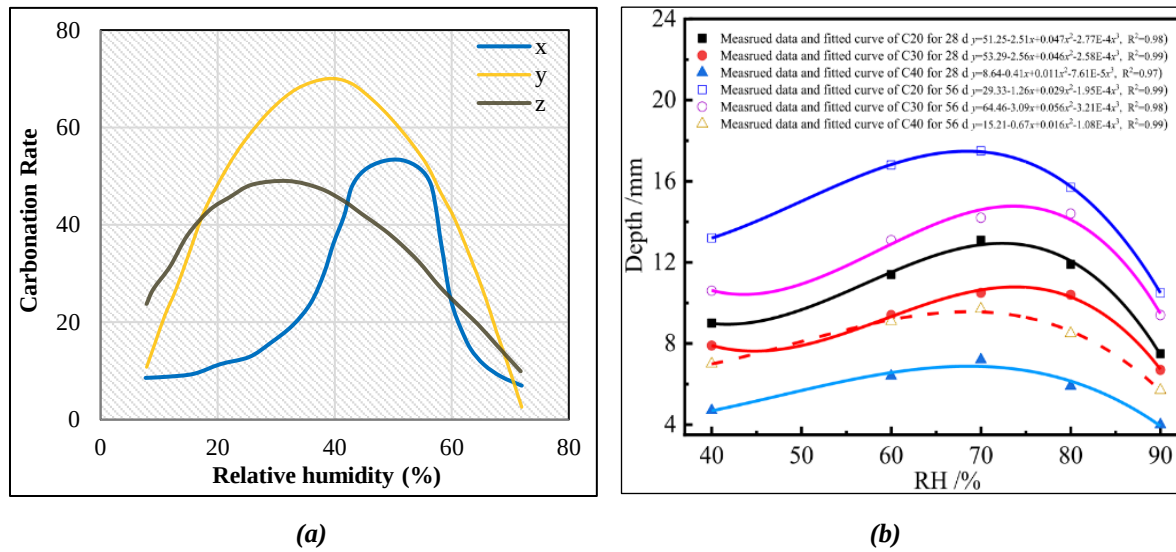


Fig. 7 – (a) (X [21], Y [22], Z [23]) and (b) [73] shows the relation between the relative humidity and carbonation efficiency

3.2.5 Effect of temperature

The process of carbonation is significantly impacted by temperature. Concrete carbonation is a chemical reaction process. The correlation between temperature and the chemical reaction coefficient is described by an exponential function based on the Arrhenius law (equation 6).

$$d_c = a + b \exp(T/t_1) \quad (6)$$

Where d_c is the carbonation depth in mm, a , b , and t_1 are the fitted constants, and T is the temperature of carbonation in Kelvin. Fig. 8(a) shows the relationship between the temperature and the carbonation depth. It is evident that the carbonation depth increases as the temperature of carbonation increases [73].

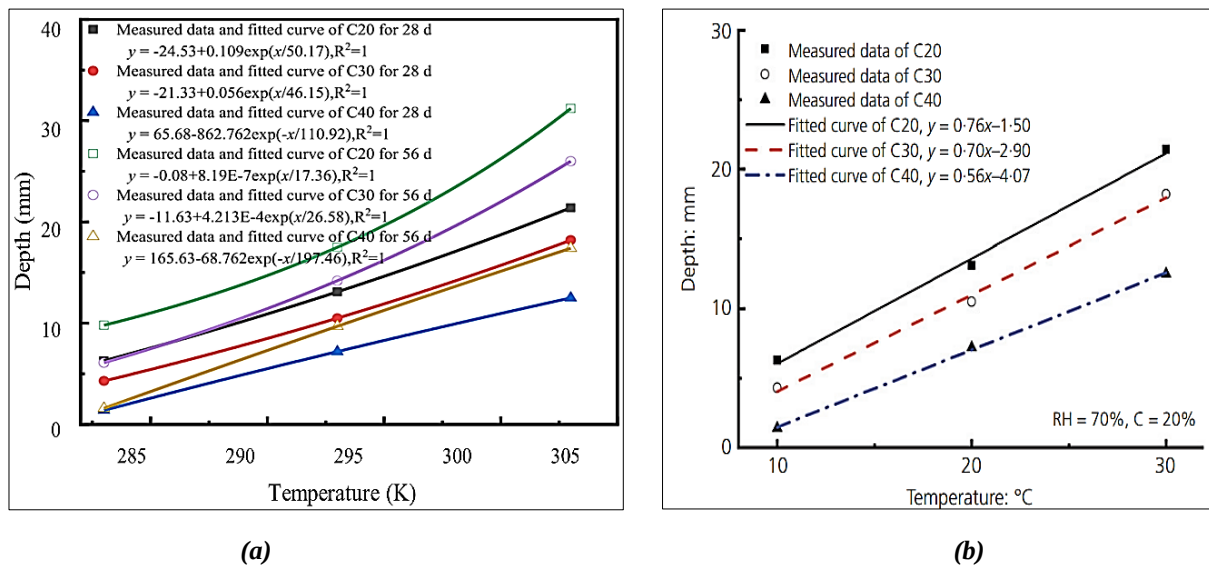


Fig. 8 – (a) [73], (b) [74] Relation between temperature and carbonation depth

Liu et al. [74] found that the relation between concrete carbonation and temperature could be characterised by a linear function in which carbonation depth increases linearly with the increase in temperature, as presented in Fig. 8(b). Regardless

of age, Lu et al. [75] discovered that the carbonation depth rises with temperature. For instance, following 28 days of CO₂ curing at 5 °C, 20 °C, and 50 °C, the sample carbonation depth was 12.2 mm, 18.5 mm, and 21.8 mm, respectively. Moreover, samples that were cured at 50 °C reached a carbonation depth roughly twice as deep as those that were cured at 5 °C, indicating that a faster carbonation rate was produced by a higher curing temperature. The coefficients of reaction and transmission increase with temperature. Consequently, the carbonation and reaction kinetics levels in concrete samples are more significant [71].

3.2.6 Effect of recycled concrete aggregates (RCA)

Recycled concrete aggregate (RCA) can include bituminous materials, ceramics, unbound aggregate, and floating particles. Different physical and mechanical properties of RCAs impact the concrete performance. Aggregate replacement ratio, w/c ratio, adhered mortar, and aggregate size have an adverse impact on the carbonation resistance of concrete [76, 77]. The replacement ratio of RCA on the carbonation resistance of concretes without and with SCMs revealed that the carbonation depth rises 1.06 to 2 times when coarse aggregates are replaced in comparison to normal concrete [78–80]. Recycled aggregate, if used as a fine aggregate, increases the carbonation depths up to 3.75 to 12.25 times more than the concrete with natural aggregate [81–83]. Zhan et al. [65] investigated the impact of RCA on the carbonation process using particle sizes of 5–10 mm, 10–14 mm, and 14–20 mm. After one hour of CO₂ curing, it was observed that smaller particle sizes and a greater specific surface area allow RCA to absorb more CO₂ during the CO₂ curing process. For RCA with a particle size of 5–10 mm, the carbonation percentage could reach up to 56%, but for RCA with a particle size of 14–20 mm, it was only 37%. The implication is that smaller RCAs have a vast potential for CO₂ capture.

3.2.7 Effect of supplementary cementitious materials (SCMs)

The huge amount of CO₂ emissions linked to cement production has reduced cement production and utilization in concrete construction. Supplementary cementitious materials (such as fly ash, silica fume, slag, and metakaolin), due to their very fine size and pozzolanic properties, play an essential role in the partial substitution of cement [84]. Zhang et al. [19] investigated the CO₂ uptake of the OPC and fly ash-substituted OPC. In relation to the mass of cement, it was found that pastes containing a higher fly ash component absorbed more CO₂. The reaction rate was initially high, but it significantly slowed down after four hours. When cement is substituted by fly ash at the same water-to-binder ratio, the effective w/c of the paste increases. The distance between cement grains enlarges due to fly ash particles and free water, providing higher possibilities for the reaction with CO₂ because carbonation occurs in aqueous conditions. It is clear from Fig. 9 that the partial substitution of cement with fly ash increases the carbon uptake.

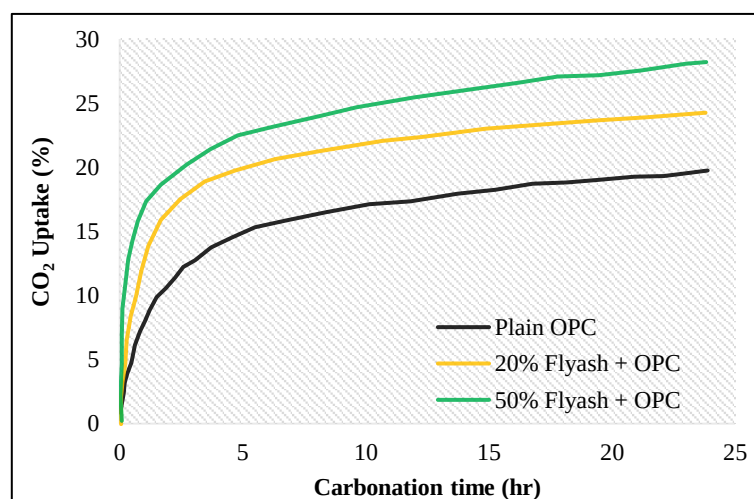


Fig. 9 – CO₂ uptake mass curves of plain cement paste and with fly ash substitution [19]

In the case of high-volume fly ash concrete (HVFA), the CH is consumed by the pozzolanic hydration reaction of fly ash [20]. As a result, the carbonation front moves inwards faster due to less CH availability. The lower CH availability will result in more C–S–H carbonation [39].

Singh and Singh [85] noted that, at all curing ages and for all exposure times, the carbonation depths of self-compacting concrete (SCC) rise with an increase in fly ash content. When compared to low-volume fly ash (LVFA) based mixes, carbonation resistance for high-volume fly ash (HVFA) based mixes (incorporating RCA) has been observed to be drastically reduced. Fly ash with high calcium content (more than 20 percent by mass) is also a strong contender for CO₂ sequestration, whereas low-calcium fly ash only has a modest capability for carbonation. However, it was observed that the CaO content has less impact on the carbonation efficiency of the materials, suggesting that the primary determinants of the carbonation efficiency are the material microstructure and particle sizes, as well as the accelerated carbonation conditions [86]. Papadakis et al. [87] reported increased carbonation depth when cement was partially replaced with silica fume (SF) and low and high-calcium fly ash. Nevertheless, the carbonation depth drops when low and high-calcium fly ash and silica fume are used instead of coarse particles. It was found that the carbonation depth increases with increased cement replacement by SCMs and decreases with increased aggregate replacement by SCMs like silica fume, and low and high-calcium fly ash. High-Ca fly ash has the lowest carbonation depth, followed by low-Ca fly ash, while silica fume exhibits the maximum carbonation depth. K. Byfors [88] reported that adding 10-15% silica fume does not influence carbonation when compared to the OPC control. However, the addition of 15-40% pulverised fly ash showed a higher rate of carbonation. When silica fume and fly ash are used simultaneously, the carbonation depth was found to be lower because of the lower porosity [89]. These findings are in agreement with the findings reported in [90]. Zhang et al. suggested that 9.8 percent limestone filler in Portland limestone cement has no significant effect on compressive strength and carbonation depth, but using 25% ground granulated blast furnace slag (GGBFS) in addition to limestone filler as a cement replacement increases the carbonated area of the specimens [91].

3.2.8 Effect of Concrete Grade

In concrete with lesser strength, the carbonation rate appears to increase with increasing CO₂ concentration; however, this effect is less pronounced in concrete with greater strength of more than 40 MPa due to the low rate of CO₂ diffusion [92]. Northwood performed the carbonation tests on the concrete samples after 7-42 days of exposure. It was observed that lower grades of concrete showed more carbonation depth and vice versa. The carbonation rate constant k (mm/yr^{0.5}) was determined for different cube strengths according to equation 7.

$$d = kt^{0.5} \quad (7)$$

Where d is the carbonation depth in ‘mm’ and t is the exposure time in years. The calculated k values are given in Table 1 [93].

Table 1 – The relation between the concrete grade and the carbonation rate constant

w/c	Cube Strength (MPa)	Carbonation depth (mm) after various days of exposure						Carbonation rate constant k (mm/yr ^{0.5})
		7 days	14 days	21 days	28 days	35 days	42 days	
0.55	27	0	1	2.2	3.0	3.7	5.3	10.89
0.60	26.5	0	1	2.8	3.3	4.5	5.5	11.9
0.65	23.5	0	1	3.3	3.8	5.1	7.1	13.7
0.70	20.5	0	1	3.9	4.3	6.1	7.5	15.5
0.75	18.5	0	1	4.1	4.7	6.3	8.5	16.9

From Table 1, it is clear that the rate of carbonation increases as the grade of concrete decreases. Since the pore structure of concrete is greatly influenced by both carbonation and compressive strength, the depth of carbonation falls as compressive strength increases [94].

3.2.9 Effect of waste Rubber tire

Using waste tire rubber chips to replace part of the natural aggregates reduces the consumption of natural aggregates in the concrete industry and helps resolve excessive waste tire accumulation. With different sizes and proportions of rubber aggregates, different authors have come up with different results. Yu et al. [95] studied the effect of the size and dosage of

waste rubber on the carbonation resistance of the concrete. The experimental results indicated that the smaller the particle size of the rubber aggregates, the more the anti-carbonation properties, and the less the chances of carbonation. Among the dosages of 5%, 10%, and 15% of the waste tire, it was found that 10% is the optimum dosage for the highest anti-carbonation property of the concrete. When using shredded waste tire as fine aggregate in the concrete, it was observed that with concrete with up to 30% tire addition, the carbonation depth decreases, and with 40% to 60% tire rubber addition, the carbonation depth again starts increasing, matching or exceeding that of the control sample [96]. Some studies found that, as the percentage of tire rubber in the concrete increases, there is a proportional increase in the carbonation depth [97–99]. Garros replaced 25% fine and coarse aggregates in concrete with rubber aggregates and found that carbonation depth decreased in both cases. Still, the effect is more noticeable in the case of fine aggregate replacement [100]. Liu et al. [101] prepared a hybrid fiber rubber concrete (HFRC) using basalt fibers, polypropylene fibers, and rubber particles (to replace fine sand). It was observed that the carbonation resistance of HFRC is ideal when the content of basalt fiber, polypropylene fiber, and rubber is 0.2%, 0.15%, and 5%, respectively.

3.2.10 Effect of Nanomaterials

Nowadays, nano TiO_2 (NT), nano CaCO_3 (NCa), nano SiO_2 (NS), and other nanomaterials are the most commonly used nanomaterials in binder materials. Nanomaterial integration in binder materials affects the degree of matrix carbonation response in two ways. Nanomaterials improve the specific surface area for the carbonation process, refine grain size, supply additional nucleation sites for cement hydration products, and encourage the carbonation of the matrix. However, nanomaterials limit persistent carbonation and improve the carbonation resistance of binder materials by speeding up the cement hydration reaction, increasing the rate of hydration product formation, reducing the cement matrix pore size, and further promoting the compactness of concrete. According to Moro et al. [102], as the titanium dioxide nano content rises, the carbonation of the binder materials first shows a rising trend before declining. By microregulating the hydration products, adding nanoparticles to binder materials can enhance the hydration products' orientation and crystalline size. As a result, it can potentially increase the rate of early carbonation reactions and prevent prolonged carbonation. Ramachandran et al. [103] observed the effect of nanoparticles on the carbonation resistance of four different types of M35 grade concrete mixes, including fly ash concrete (containing 40% fly ash by weight of cement), fly ash concrete modified by TiO_2 , fly ash concrete modified by CaCO_3 , and fly ash concrete mix containing an equal ratio of TiO_2 and CaCO_3 nanoparticles. The 50–80 nm-sized nanoparticles were used at two weight percent of OPC. It was found that adding nanoparticles to fly ash concrete increased its resistance to carbonation. Excellent carbonation resistance was displayed by the mix that contained both TiO_2 and CaCO_3 in equal proportions.

3.2.11 Effect of Alkali-activated Materials

The microstructural alterations in alkali-activated GGBS mortars, activated by NaOH and a sodium silicate solution with varying Na_2O /precursor dosages and $\text{SiO}_2/\text{Na}_2\text{O}$ ratios, were documented by Shi et al. [104]. The mortars were steam-cured for 48 hours and then carbonated for 56 days at 3 ± 0.2 % CO_2 at atmospheric pressure. The results indicated that increasing the activator's Na_2O content and $\text{SiO}_2/\text{Na}_2\text{O}$ ratio enhanced the carbonation resistance. Another study by Li et al. [105] on the alkali-activated GGBFS mortars showed that the materials activated with NaOH alone showed higher resistance to carbonation than the materials activated with NaOH and sodium silicate solution. Additionally, Carvalho et al. [106] evaluated the impact of varying the amounts of NaOH and Na_2SiO_3 on the mechanical performance of alkali-activated mortars prepared with municipal solid waste incinerated bottom ash as a precursor and cured under forced carbonation. Compared to carbonated mixes activated with NaOH and Na_2SiO_3 , the carbonated mixes activated with NaOH alone demonstrated higher compressive and flexural strengths. Furthermore, mechanical performance increased as the NaOH content increased. To enhance the mechanical and microstructural performance of alkali-activated electric arc furnace slag (EAFS) mortars, Kassim et al. [107] looked into the impact of utilizing various mixes of NaOH and Na_2SiO_3 for the alkaline solution. As in previous investigations, the specimen's mechanical performance, carbonation resistance, and denser microstructure were all improved by increasing the ratios of $\text{SiO}_2/\text{Na}_2\text{O}$ and Na_2O /binder.

Activated carbon is considered the greatest solid adsorbent because of its various benefits and inexpensive cost. Agricultural waste can be used as a precursor in creating activated carbon [108]. A biochar that has been chemically or physically activated is called activated carbon. It can regenerate readily because of its easily customizable pore structure and large surface area [109]. Adding biochar can increase the early strength of cement mortar regardless of w/c ratio [110]. The crucial biochar content ranges from 4 to 6 % [111]. The carbon fixation capacity of cement-based materials is increased by

effectively improving the carbon capture performance by adding porous biochar and altering the pore structure to promote transmission [112]. The tensile plane is less strong in regions where the concentration of biochar particles is more than 6.5 % because of inhomogeneity [113]. The aggregation action of the biochar particles weakens the overall strength of the concrete, resulting in local weak zones.

4 Influence of carbonation on the mechanical and durability properties of concrete

4.1 Compressive strength

The ability of concrete to withstand breaking down under the action of stresses from the compression load is expressed as its compressive strength. Significant strength gains have been noted at early ages up to 24 hours due to CO₂ curing. Compared to conventional water curing for three and seven days, the compressive strengths of cement paste specimens carbonated for four hours are higher [62]. Chi et al. found that the compressive strength of carbonated concrete samples was more than that of non-carbonated samples because of the CaCO₃ deposition in the pore structure of the carbonated samples [94]. When the recycled aggregates were first carbonated and then used, a comparable compressive strength after 28 days of moist curing was obtained [114]. The improvement in performance is ascribed to the lower porosity and an improvement in the ITZ [115]. It was found by Xiao et al. [116] that the compressive strength of carbonated coarse aggregate concrete and carbonated fine aggregate concrete was 13.1% and 16.5% greater than that of the concrete made with identical amounts of mixed ratio of non-carbonated recycled aggregates, respectively. The main reasons are that the adhering mortar on both recycled fine aggregate (RFA) and recycled coarse aggregate (RCA) is strengthened by the CO₂ treatment. However, the strength values of carbonated concrete decreased after 180 days because of the limited strength of the recycled aggregates, which the carbonation treatment cannot further strengthen [117]. Compared to moist curing, the CO₂ cured process improved the compressive strength of concrete samples by 15.4%, 18.9%, and 28.8% after 6, 12, and 24 hours, respectively [118] as shown in Fig. 10(a). Interestingly, a strength greater than 20 MPa was achieved after only two hr of CO₂ curing at 20 °C and 50 °C, indicating that CO₂ curing could significantly reduce the curing time [119], as depicted in Fig. 10(b). Similar results were reported by Shi et al. [17].

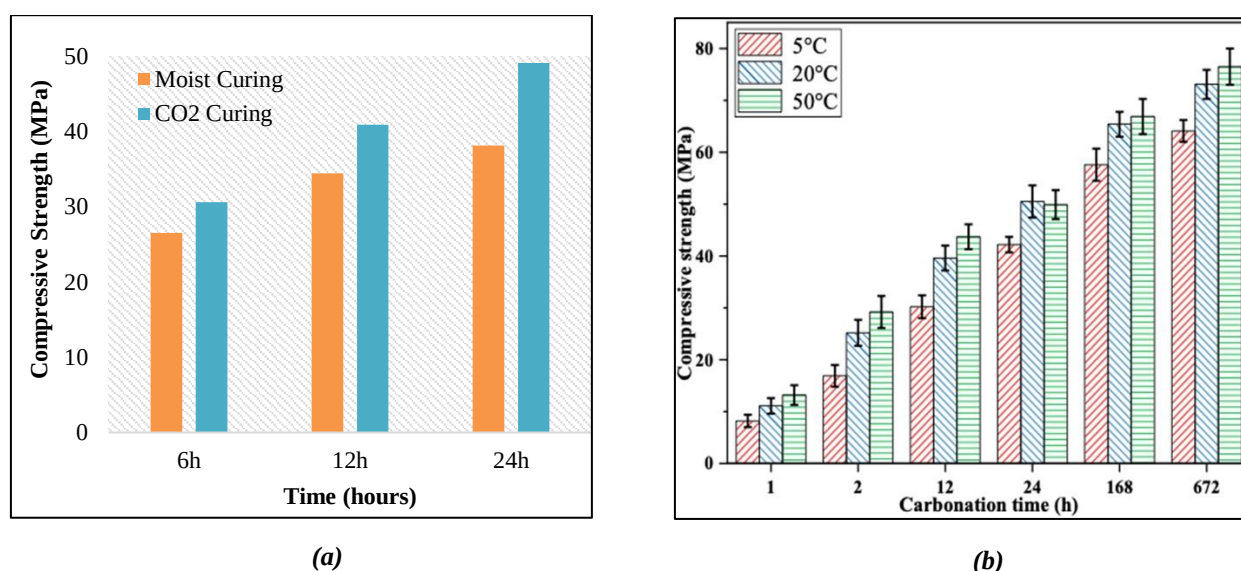


Fig. 10 – Comparison of Compressive strength between (a) moist and CO₂ curing[118] (b) carbonation time [119]

Carbonation test results by Monkman et al. [120] show that CO₂ curing significantly increases strength at an early age, up to 24 hours. A concrete sample that was CO₂-cured for 2 hours showed a compressive strength of roughly 90.7% of the sample that was CO₂-cured for 24 hours. After two hours of moist-curing, the compressive strengths of paste carbonated for two hours at 5 °C and 20 °C were almost three and nearly four times higher, respectively. Zhan et al. [121] employed CO₂ curing on the freshly prepared OPC pastes with a w/c ratio of 20.18 and discovered that the compressive strength of the paste

after 2 hours of carbonation curing achieved a compressive strength greater than 66.1% of that of the sample carbonated for 24 hours

4.2 Split tensile strength

The ability of concrete to withstand tensile stresses is indirectly indicated by splitting tensile strength. An increase in w/c and carbonation led to an increase in the splitting tensile strength of concrete. The rationale is that the concrete with a greater w/c ratio has a faster rate of carbonation, which means that at the same carbonation time, the carbonation-induced improvement in mechanical properties becomes more substantial [122]. Splitting tensile strength tests on self-compacting concrete (SCC) mixes with corresponding water-to-powder proportions revealed that increased water-to-powder resulted in strength reduction. The splitting tensile strength of non-carbonated samples was found to be less than that of carbonated samples [123]. When recycled aggregates are used in place of natural aggregates, the split tensile strength decreases. However, when carbonated recycled aggregates are used, the split tensile strength of the concrete increases [124]. Fig. 11 shows the comparison of the split tensile strengths of natural (NAC), recycled (RAC), and carbonated recycled aggregate concrete (CRAC).

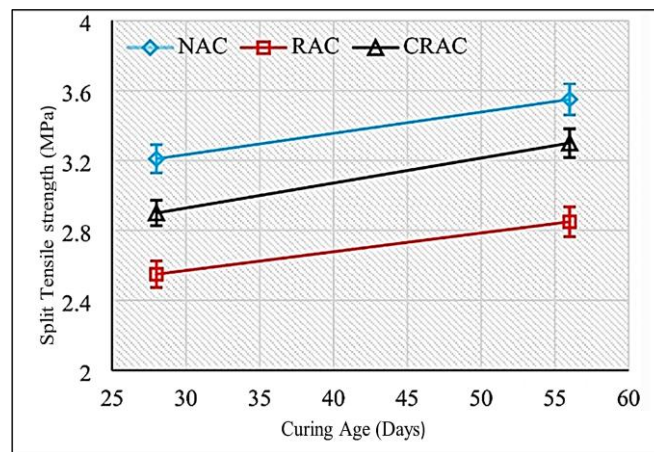


Fig. 11 – Split tensile strength test results [124]

When the quantity of recycled aggregate increases, the strength of aggregate-mortar ITZs decreases, therefore reducing the split tensile strength.

4.3 Porosity

Mercury porosities were measured by Hyvert et al. [70] on three carbonated mortars. It was observed that the porosity of the carbonated zone of the mortars decreases with the increase in CO_2 pressure, which reduces the diffusivity of the CO_2 gas. The smaller dimension pores also get clogged due to carbonation as a result of calcite formation [125]. In Portland cement paste, total porosity is reported to decrease during carbonation; however, it is observed to rise in blended cement paste. It has been found that the pores become coarser after carbonation [26, 126], but some researchers found that the pore structure becomes finer after carbonation [44, 127]. The water and gas permeability of Portland and blended cement paste has been found to reduce significantly after accelerated carbonation [44, 127, 128]. Moreover, some studies found that the combination of carbonation and capillary pore coarsening results in a decrease in the overall porosity of plain cement pastes [129, 130]. Fig. 12(a) shows the effect of carbonation on porosity.

After carbonation, the measured water permeability dramatically drops. In comparison to M45 and MS45, the water permeability of M55 and MS55 decreases more, suggesting that saturation carbonation has a more substantial impact on the pore structure of mortars with higher water/binder (w/b) ratios (Fig. 12(b)). The reduction in water permeability is attributed to the pore structure refining during carbonation. All carbonated mortars showed a 41% reduction in surface relaxivity after carbonation, suggesting that carbonation significantly alters the properties of pore walls [131]. Carbonation of the concrete reduces the porosity accessible to water, as 5% to 12% of the open porosity is closed. This reduction in porosity after carbonation makes concrete more compact [122].

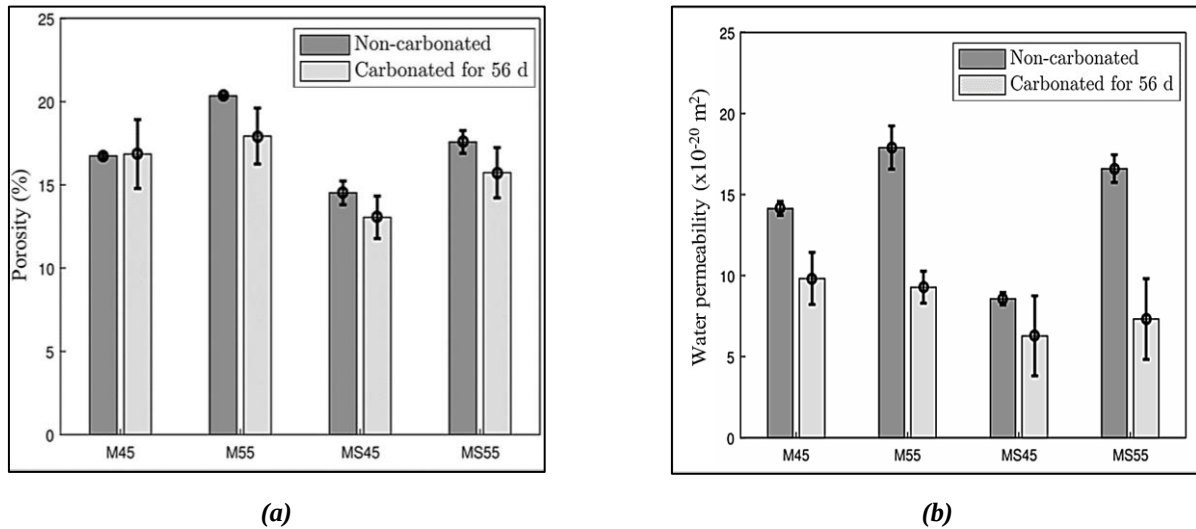


Fig. 12 – Evolution of (a) porosity and (b) water permeability after saturated carbonation [131]

(Here, ‘M’ denotes mortar, the number denotes water binder ratio of 0.45 and 0.55, and ‘S’ denotes 10 % silica fume replacement.)

4.4 Chloride Penetration

The Rapid Chloride Permeability Test (RCPT), following ASTM C1202 standards, is performed to quantify the charge (coulombs) transmitted. This measurement serves as an indicator of concrete resistance to ion migration and penetration when subjected to various curing conditions. Basu et al. [123] conducted the RCPT testing to study the concrete permeability of sandstone-modified self-compacting concrete. A correlation between the charge transmitted through the specimens and the depth of carbonation indicated that, in comparison to carbonated samples (CS), more charge was transmitted through the non-carbonated samples (NCS). Boyd et al. [132] found that at both early and later stages, the carbonated samples allowed much less electrical charge to flow through than the steam-cured specimens. Fig. 13 shows the comparison of total electrical charges passed through concrete samples at 2 days and 28 days of steam and carbonation curing. The decreased ion penetration could be attributed to the increased packing density produced due to the precipitation of CaCO_3 .

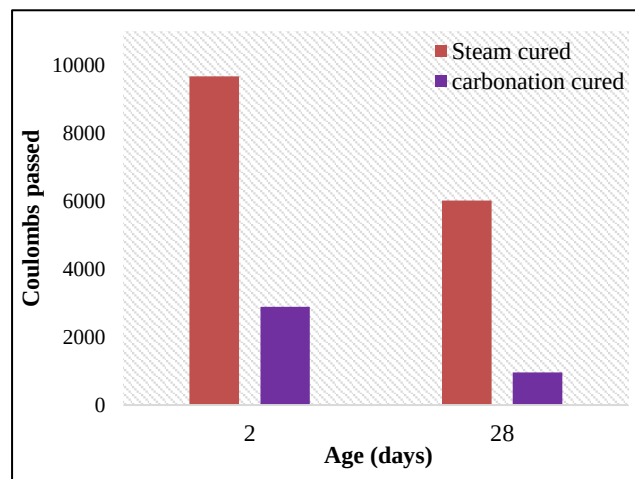


Fig. 13 – Accumulative electrical charge during RCP testing for concretes with different curing schedules (modified from [132])

4.5 Electrical conductivity

The conductivity in concrete is due to the presence of water in the capillary pores, which contain dissolved salts and act as an electrolyte [94]. The bulk electrical conductivity of the concrete specimen is determined as per ASTM C1760-12.

Electrical current flowing through a saturated concrete sample is measured in this test with a 60 V applied voltage across the specimen ends. One minute after the voltage is initially applied, the current is measured. The calculation of electrical conductivity is done using equation 8.

$$\sigma = a \frac{I_1 L}{V D^2} \quad (8)$$

Where, σ is the bulk electrical conductivity (mS/m), I_1 is the current at 1 min (mA), V is the applied voltage (V), L is the average length of the specimen (mm), D is the average diameter of the specimen (mm), and a is the conversion factor, 1273.2. More electrical resistivity indicates well-hydrated concrete. As per the results of Boyd et al. [132], carbonated concrete samples showed greater surface resistivity than steam-treated samples, seemingly due to their less porous surfaces. The lower permeability is attributed to the increase in the solid volume by 11% in the conversion of $\text{Ca}(\text{OH})_2$ to CaCO_3 [43]. When water is sprayed in addition to the carbonation, the electrical resistivity of the concrete sample increases. After carbonation, the water spray encourages greater hydration, which increases strength and improves durability. The electrical conductivity of recycled aggregate incorporated concrete increases gradually from 18 mS/m to 25 mS/m. With the increasing number of RCAs in concrete. However, the conductivity decreases with the use of carbonated RCAs [133]. From the results of Medeiros et al. [134], it was observed that the electrical resistivity of saturated carbonated specimens is higher than that of non-carbonated samples with the same water-cement ratio. The calcium carbonate formed in the carbonation occupies more space and contributes to refining the concrete pore. Consequently, the resistivity rises, and the concrete matrix becomes denser. The variation is shown in Fig. 14.

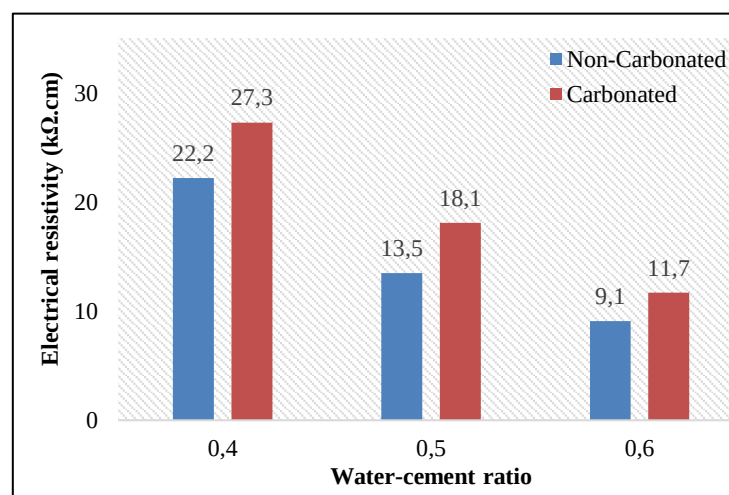


Fig. 14 – Electrical resistivity of saturated carbonated and non-carbonated specimens [134]

4.6 Sorptivity

Sorptivity serves as an indicator of moisture transport into unsaturated specimens, exhibiting sensitivity to both the type and duration of curing [135]. It is a useful indicator of the near-surface concrete's quality, which controls corrosion-related reinforcement durability [136]. The sorptivity of concrete is sensitive to the quality of the cover zone, which is greatly influenced by the extent of the curing. It was reported by Blight [137] that water absorption reduces with age and is attributed to the carbonation that occurs with age. Another study by Lampacher et al. [138] indicated that the water sorptivity of a 25-year-old concrete section was lower in the cover zone, which was carbonated. Similar results were obtained by another study [139] on the cores cut from 30-150 year old concrete structures, which show decreasing sorptivity and increasing depths of carbonation with age, despite a decrease in strength. Medeiros et al. [134] found that the capillary absorption and water-cement ratio are directly proportional; the reason is attributed to the larger quantity of pores in the higher water-cement ratio [139]. However, because of the pore refinement brought by the carbonation reaction, the carbonated samples show greater capillary absorption than the non-carbonated concretes [134], as shown in Fig. 15. Smaller pores increase the sorptivity capacity and capillary tension. The absorption by capillaries is higher for small-diameter holes. The kinetics of carbonation increase with the water/cement ratio of the concrete mix, which is directly attributed to the role played by porosity [140].

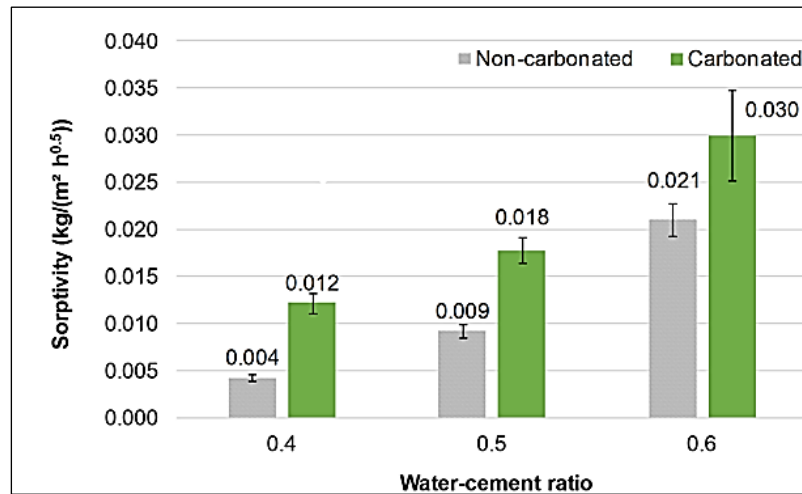


Fig. 15 – Sorptivity values for carbonated and non-carbonated specimens [134]

4.7 Shrinkage

The carbonation shrinkage is very similar to the drying shrinkage in terms of its time of occurrence. However, the mechanism is entirely different. Carbonation shrinkage is an irreversible volume change that occurs due to the chemical reaction with atmospheric CO₂ [141]. Using a horizontal length comparator as specified by AS 1012.13 (2015), the embedded shrinkage pins measure the change in length caused by concrete shrinkage from end to end. The rate of carbonation determines the rate of shrinkage due to carbonation. Y. Houst observed [142] that the carbonation shrinkage increases with the time of carbonation. The carbonation shrinkage of hydrated cement paste decreases with an increase in relative humidity, but an increase has been found in the shrinkage when the RH ranges from 60 to 75%. After that, shrinkage again decreases (Fig. 16).

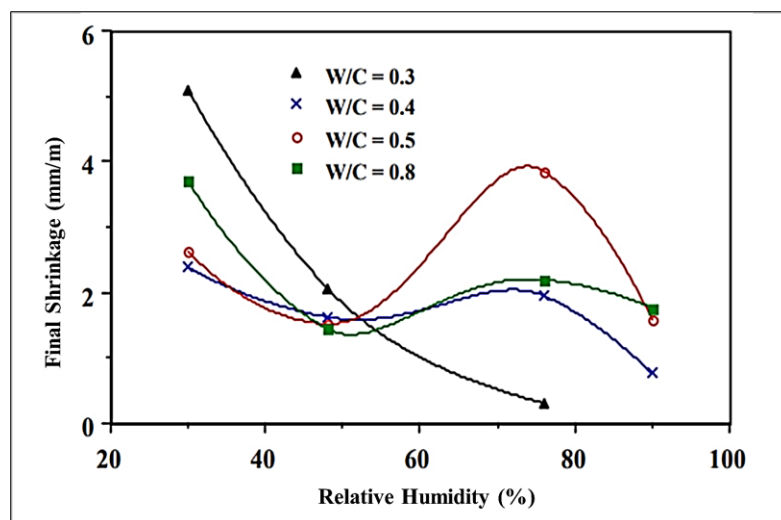


Fig. 16 – Carbonation shrinkage vs relative humidity [142]

Hailong et al. observed carbonation expansion rather than shrinkage when alkalis, such as sodium salts, are added to the OPC. Also, the shrinkage magnitude continues to increase at constant mass change [141]. In the case of high-performance concrete (HPC), it was observed that the carbonation shrinkage increases with the increase in the w/c ratio. At a low w/c ratio of 0.28 and with a 10% silica fume content, whole carbonation shrinkage was eliminated because of the consumption of calcium hydroxide in the HPC. No carbonation shrinkage was observed within the four years of carbonation [143]. Table 2 summarises the effect of carbonation on the mechanical and durability properties of concrete.

Table 2 – Summary of carbonation effects on the mechanical and durability properties of concrete

Property	Effect of Carbonation
Compressive Strength	↑ (Carbonation curing significantly increases compressive strength at early ages, up to 28% higher than non-carbonated)
Split Tensile Strength	↑ (Enhanced, especially with carbonated recycled aggregates)
Porosity	↓ (Pores refine or partially clog; concrete becomes denser)
Chloride Penetration	↓ (Reduced ion migration due to CaCO_3 precipitation)
Electrical Resistivity	↑ (Denser matrix, lower permeability)
Sorptivity	↑ (Capillary absorption increases due to finer pores)
Shrinkage	↑ / ↓ (Depends on w/c ratio, RH, silica fume; possible expansion with alkalis)

5 Natural vs Accelerated Carbonation

As mentioned earlier, the typical range of CO_2 levels in the atmosphere has been rising over the past two centuries, exceeding 400 ppm or 0.04% since 2015, and continues to increase. Natural carbonation of concrete would be the most realistic means of examining the performance of concretes under CO_2 exposure, but the natural process is too slow and takes a very long time to exhibit a significant effect. So, accelerated carbonation studies are commonly used in the laboratory to rapidly evaluate concretes. Over the past decades, the accelerated method of carbonation has been standardized by using high CO_2 concentrations, such as 4% CO_2 maintained at 20 °C and 55% RH [84]. The standard CEN/TS 12390-10 also recommends a “natural test” laboratory method intended to simulate a natural exposure environment by employing a typical natural CO_2 concentration of 0.035% at 20 °C and 65% RH [144, 145]. RILEM [146] provides guidelines for carbonation testing on real structures and laboratory tests. Various experiments reported in the literature have used widely varying CO_2 levels for accelerated tests. These include 1% CO_2 [147], 4% CO_2 [74], [148, 149], 5% CO_2 [150, 151]. Loo et al. [92] used 7%, 12%, and 18% CO_2 concentrations in their accelerated carbonation experiments, while others have applied a 20% CO_2 concentration [152, 153]. Some studies have found a clear association between the accelerated and natural carbonation, and it is suggested that 7–15 days of accelerated carbonation are equivalent to a year of natural carbonation. Additionally, Dhir et al. found that a week-long accelerated carbonation test corresponds to roughly fifteen months of natural exposure [149]. Despite the use of results from accelerated carbonation tests to predict long-term carbonation depths, there is no consensus on the method for extrapolating these accelerated test results to real-world performance [150, 154]. The complexity of this issue arises from the differences in mass transport properties between laboratory specimens and structural elements, as well as the variance in CO_2 concentrations between accelerated testing and natural exposure conditions. According to Fick’s first law, the carbonation depth is proportional to the square root of the CO_2 concentration. However, some studies [155, 156] indicated that other factors involved in the kinetics of carbonation reactions may influence this relationship, potentially rendering the proportionality invalid [157].

Two major groups of factors influence the carbonation of concrete: environmental factors and intrinsic properties of the concrete itself. Environmental factors encompass the occurrence of dry-wet cycles [158], relative humidity, and CO_2 concentration [159]. Intrinsic factors are primarily associated with the porous structure of the concrete, which affects its mass transport properties, and the quantity of substances available for conversion into carbonates. The carbonation coefficient encapsulates several parameters typically considered constant: CO_2 concentration, the amount of CO_2 needed to carbonate a unit volume of concrete, and the diffusion coefficient for CO_2 through carbonated concrete. However, under real exposure conditions, CO_2 concentration varies over time [160]. The diffusion coefficient is influenced by relative humidity and dry-wet cycles, and the amount of CO_2 required to carbonate a unit volume of concrete depends on the available CO_2 [161]. At typical CO_2 concentrations, only the hydrated cement compounds react with CO_2 , whereas at higher CO_2 concentrations, both hydrated and anhydrous cement compounds react [162].

Sanjuan et al. [150] conducted natural and accelerated carbonation tests on concretes incorporating CEM I 42.5R, CEM I 52.5N, and Class C & F fly ash with w/cm ratios ranging from 0.33 to 0.69. Their findings indicated that CO₂ concentrations of 5% and 100% resulted in 5 and 40 times higher carbonation rates, respectively, compared to natural exposure (0.03% CO₂). Similarly, Dhir et al. [163] and Ho & Lewis [164] found that one week of accelerated carbonation approximates 15 months of natural carbonation.

Long-term carbonation studies by Oyinkanola et al. [165] examined the impact of RH and CO₂ concentration over 50 years under sheltered conditions. Their study compared three cases: Case 1: Identical CO₂ concentration and RH resulted in comparable carbonation depths. Case 2: Higher CO₂ concentration at the same RH led to greater carbonation depth. Case 3: At constant CO₂ concentration, carbonation was maximized within the optimal RH range of 50–70%.

Similarly, Seo et al. [166] studied carbonation in alkali-activated slag (AAS) using six activators (NaOH, Na₂SiO₃, Na₂CO₃, Na₂SO₄, CaO, MgO). Their findings demonstrated that 28 days of accelerated carbonation (1% CO₂) equalled 2 years of natural carbonation. Table 3 summarises the findings on natural and accelerated carbonation.

Table 3 – Key findings on accelerated and natural carbonation.

Ref	Key Findings on Carbonation	Comparison Between Natural & Accelerated Carbonation
[167]	Higher CO ₂ leads to faster carbonation but an altered microstructure. Correction factors were applied.	Accelerated carbonation tests underestimate natural carbonation depth.
[168]	Developed a linear correlation model between natural and accelerated carbonation.	Found high correlation ($R^2 = 0.90+$), enabling accurate long-term predictions.
[169]	Studied high-volume fly ash (HVFA) mortar. Observed coarser pore structures under accelerated conditions.	Correction factor ~9.9 for high-volume fly ash mixtures.
[170]	Developed a carbonation model for fly ash, slag, and limestone-calcined clay concrete.	Found poor direct correlation but proposed an improved A-to-N model for accurate prediction.
[171]	Studied lime-based materials under different CO ₂ conditions. Larger calcite crystals formed under accelerated carbonation.	Accelerated carbonation at 5% CO ₂ for 7 days equalled natural carbonation of 3 years.
[172]	Investigated carbonation in concretes with shrinkage modifiers and internal curing.	Found good correlation, but fly ash mixes had higher carbonation depths.
[173]	GGBS-based concretes have lower carbonation resistance than OPC concretes.	Higher GGBS content led to higher carbonation depths, but trends were not identical.

5.1 Correlation Between Natural and Accelerated Carbonation

Numerous studies confirm that accelerated carbonation tests can predict long-term natural carbonation, but correction factors are needed to adjust for environmental variability. The correlation between natural carbonation coefficient (K_n) and accelerated carbonation coefficient (K_a) is typically established using linear regression analysis. Most studies report a strong linear relationship ($R^2 > 0.90$) [168], validating its use in predictive carbonation models. However, variables such as CO₂ concentration, humidity fluctuations, and supplementary cementitious materials (SCMs) introduce uncertainties that require further refinement.

Forsdyke & Lees [167] observed that accelerated carbonation underestimates natural carbonation depth, primarily due to differences in environmental exposure. Neves et al. [168] developed a linear correlation model and determined that K_a/K_n ranges from 9.8–15.0, meaning accelerated carbonation progresses 9.8–15 times faster than natural carbonation. Similarly, Heede et al. [169] found that high-volume fly ash (HVFA) concrete showed higher carbonation depth under accelerated conditions, necessitating a correction factor 9.9 for accurate predictions.

The effect of CO₂ concentration on carbonation depth is also significant. Studies show that above 10% CO₂, carbonation accelerates, but beyond 20%, the rate stabilizes due to saturation effects. Silva et al. [171] observed that 7 days of accelerated

carbonation at 5% CO₂ corresponded to 3 years of natural exposure, confirming the viability of controlled acceleration methods. However, GGBS-based concretes showed greater carbonation susceptibility, requiring adjusted correction factors.

5.2 Carbonation Depth vs. Carbonation Coefficient Relationship

The relationship between carbonation depth (X) and carbonation coefficient (K) is often assumed to follow Fick's Law, where carbonation depth is proportional to the square root of time ($x = K\sqrt{t}$). However, the actual carbonation depth ratio (X_a/X_n) observed in experimental studies is significantly lower than the carbonation coefficient ratio (K_a/K_n). Table 4 summarises the carbonation coefficient ratios and carbonation depth ratios.

Table 4 – Carbonation coefficient ratio (K_a/K_n) and carbonation depth ratio (X_a/X_n)

Ref	Natural Carbonation Coefficient (K_n) (mm/ $\sqrt{\text{year}}$)	Accelerated Carbonation Coefficient (K_a) (mm/ $\sqrt{\text{year}}$)	(K_a/K_n)	Natural Carbonation Depth (X_n) (mm)	Accelerated Carbonation Depth (X_a) (mm)	X_a/X_n
[173]	2.7	27.0	10.0	4.0 - 8.0	10.3 - 25.0	2.6 - 3.1
[171]	2.1	20.5	9.8	3.5 - 6.2	9.5 - 18.0	2.7 - 3.4
[168]	3.1	26 - 30.5	9.8 - 15	2.1 - 4.5	20.5 - 45.2	9.9 - 15.0
[172]	3.4	34.0	10.0	6.65 - 8.45	14.0 - 18.8	2.1 - 2.5
[169]	4.2	41.6	9.9	5.2 - 7.0	22.5 - 38.0	4.3 - 5.4
[167]	2.5 - 3.0	7.5 - 15.0	3.0 - 5.0	3.0 - 6.0	14.0 - 30.0	4.7 - 5.5

While many studies report K_a/K_n values between 9.9 and 15.0, the corresponding carbonation depth ratio is typically between 2.5 and 5, indicating that carbonation depth does not scale directly with the coefficient. Fig. 17 shows the correlation between carbonation coefficient ratio (K_a/K_n) and carbonation depth ratio (X_a/X_n).

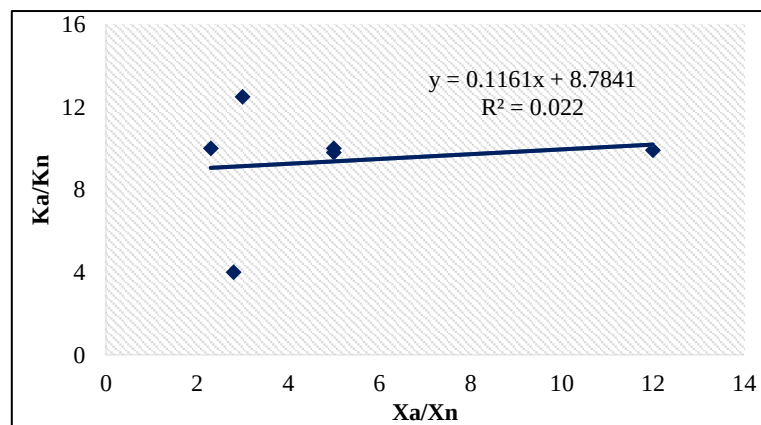


Fig. 17 – Correlation between (K_a/K_n) and (X_a/X_n)

It can be observed that a weak correlation exists between the two coefficients, with R^2 of 0.022. This discrepancy arises due to several factors. Firstly, the assumption of perfect CO₂ diffusion in Fick's Law does not fully capture the effects of moisture, porosity, and hydration reactions, which slow carbonation, especially in natural conditions. Secondly, higher CO₂ concentrations in accelerated tests (>5%) enhance carbonation, but this effect saturates beyond a certain depth, preventing a direct 10x increase in carbonation depth. Additionally, environmental factors such as fluctuating humidity and wet-dry cycles in natural carbonation slow the process compared to the controlled conditions of accelerated carbonation. Furthermore, concrete composition, particularly fly ash and GGBS-based mixes, influences the carbonation rate differently in natural vs. accelerated conditions, leading to variations in observed depth ratios. While carbonation depth (X) is proportional to the square root of time ($x = K\sqrt{t}$), experimental studies show that $X_a/X_n < K_a/K_n$. Despite K_a/K_n values between 9.9 and 15.0, actual X_a/X_n ratios range between 2.5 and 5, indicating that carbonation depth does not scale linearly with K_a .

5.3 practical implications of K_a/K_n vs X_a/X_n ratios

The ratios K_a/K_n and X_a/X_n provide a practical means of evaluating the reliability of accelerated carbonation tests for predicting long-term field performance. A K_a/K_n ratio greater than unity indicates that accelerated conditions tend to overestimate carbonation rates, while lower values may underestimate field behaviour. Similarly, the X_a/X_n ratio helps in assessing the comparability of carbonation depths obtained under controlled versus natural exposure. These ratios are therefore essential for calibrating service-life models, as they highlight the limitations of directly applying laboratory results to real structures exposed to fluctuating humidity, temperature, and microclimatic conditions. By incorporating these ratios, engineers and researchers can better adjust accelerated test outcomes to align with field performance, ultimately improving the accuracy of durability forecasts and developing maintenance strategies.

6 Case Studies

6.1 Carbonation Analysis of the Itaipu Dam

The Itaipu Dam, a monumental engineering project, required vast quantities of raw materials, primarily concrete, supplemented with pozzolanic materials such as fly ash and blast furnace slag to enhance both technical and financial efficiency [174]. The dam features a gravity-type concrete structure, constructed without reinforcement, maximizing the benefits of carbonation. Spanning 7,919 meters with a maximum height of 196 meters, the Itaipu Dam comprises multiple sections, including a main dam, right-side dam, connection blocks, and a deviation structure, as illustrated in Fig. 18.



Fig. 18 – Layout of Itaipu Dam

To assess carbonation depth, CO_2 uptake, and moisture distribution, 155 concrete core samples were extracted from various dam levels, ranging from the base (40 m) to the crest (225 m). These levels coincided with dam galleries, providing access for sampling. The internal moisture content was measured using a Hygropin device, with 48 measurements conducted at eight locations, each with at least six replications.

Key Findings include: (1) Carbonation depth increased with height. The least carbonation was observed at the base of the dam, attributed to the high-strength concrete (35 MPa) and moisture levels above 90% [43]. (2) Higher carbonation was observed at greater heights, where concrete strength gradually reduced (32 MPa, 25 MPa, and 18 MPa) along with lower moisture content of 75%. (3) Maximum carbonation depth was recorded in low-strength concrete (18 MPa) at 75% moisture. The CO_2 sequestration potential of the dam was estimated over a 35-year exposure period. With a total surface area of $2.75 \times 10^6 \text{ m}^2$, the carbonated concrete volume was calculated at $118,250 \text{ m}^3$. Based on carbonation depth readings from core samples, the total absorbed CO_2 was 13,384 tons.

6.2 Impact of Protective Render Coat (PRC) on Carbonation in a 100-Year-Old Concrete Bridge

This case study examines the effectiveness of a cement-based protective render coat (PRC) applied during the construction of a 100-year-old concrete bridge in Slovakia, located between the towns of Sládkovičovo and Krásno nad Kysucou [175]. The study aimed to assess carbonation depth and CO_2 diffusion resistance of PRC by conducting diagnostic

tests on the bridge structure. A drilling instrument was used to extract 16 mm diameter exploratory boreholes, allowing for preliminary carbonation depth assessment. Carbonation depth was evaluated using the phenolphthalein indicator, which revealed that the surface protected by PRC exhibited minimal carbonation (≤ 2 mm), with deeper penetration (up to 15 mm) only in cracked areas. In contrast, exposed concrete surfaces (without PRC) exhibited carbonation depths of up to 50 mm, leading to concrete spalling.

Key Findings include; (1) PRC significantly reduced carbonation depth, limiting CO_2 diffusion into the underlying concrete. (2) Carbonation products accumulated in the PRC layer, effectively forming an anti-carbonation barrier by reducing pore space. (3) Carbonation depth varied negligibly between the surface and interior layers, except in areas where PRC adhesion was imperfect, leading to localized fissures. (4) After 100 years of exposure, carbonation depth beneath the PRC remained less than 10 mm, confirming its long-term protective performance. Figure 19 shows the PRC layer on the 100-year-old concrete specimen, where a minor fissure was observed at the PRC-concrete interface. This slight detachment led to a localized increase in carbonation depth, but overall protection remained highly effective.

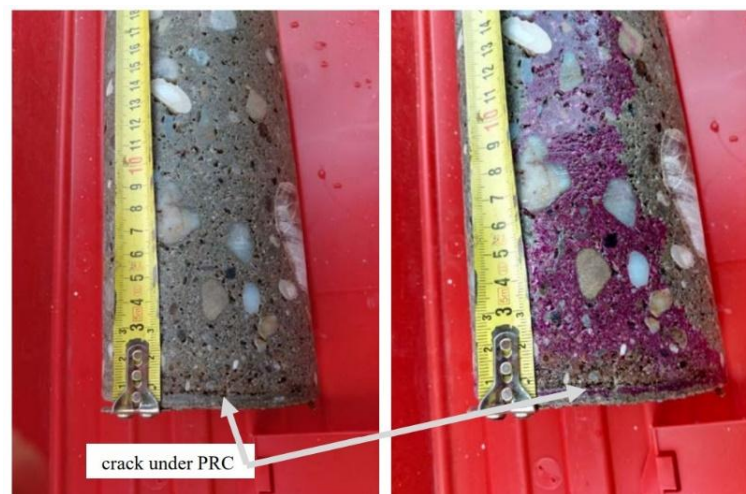


Fig. 19 – 100-year-old concrete sample protected with PRC

This case study highlights the strong correlation between carbonation resistance and the low permeability of the PRC layer. When all conditions align optimally, including high-quality PRC, absence of concrete defects, and a compact PRC-concrete interface, the carbonation depth remains exceptionally low, even after a century of service life. These findings demonstrate that PRC serves as an efficient, simple, and durable anti-carbonation barrier, effectively protecting embedded reinforcement and prolonging the lifespan of concrete structures. Table 5 presents the comparative summary between the two case studies.

Table 5 – Comparative summary between the two case studies.

Aspect	Itaipu Dam	Century-old Bridges
Age/Exposure	35 years; exposed to a natural tropical climate.	100 years; exposed to varied natural conditions.
Concrete Strength	High-strength mass concrete.	Low-to-moderate strength structural concrete.
Carbonation Depth	Limited penetration due to dense matrix and high moisture content.	Greater depth was observed, especially in areas with dry exposure and higher porosity.
Durability Impact	Low carbonation depth reduced the risk of reinforcement corrosion; long-term performance was well-preserved.	Deeper carbonation increases the risk of alkalinity loss and potential reinforcement corrosion.
Protective Factors	Massive sections, low permeability, and continuous moisture reduced the carbonation rate.	Lack of protective measures and environmental variability contributed to deeper carbonation.
Practical Implications	Demonstrates the potential of concrete structures as passive CO_2 sinks while maintaining durability.	Highlights the vulnerability of older, lower-strength concretes and the importance of protective measures.

7 Conclusions

- Carbonation significantly affects the mechanical and durability properties of concrete, governed by CO₂ concentration, relative humidity (50–70% optimal), w/c ratio, binder type, porosity, and curing.
- Carbonation refines the pore structure and increases compressive strength (10–30%) in the short term, but reduces tensile and flexural strength due to C–S–H decalcification. Over time, this leads to brittleness, microcracking, and greater shrinkage, particularly in low w/c concretes.
- Early-stage pore refinement increases electrical resistivity and lowers sorptivity; however, progressive carbonation decreases chloride-binding capacity and resistivity. The combination of carbonation and capillary pore coarsening results in a decrease in the overall porosity of plain cement pastes.
- While densification improves abrasion resistance, carbonation lowers alkalinity (pH < 9), increasing reinforcement corrosion risk, especially at high RH (>80%). Fly ash and GGBS concretes show greater carbonation depth than OPC due to lower CH content.
- Accelerated carbonation offers quick insights but cannot fully replicate natural conditions; carbonation depth does not scale linearly with the carbonation coefficient.
- Case studies show that carbonation depth is strongly influenced by concrete strength, exposure conditions, and protective measures. High-strength concrete and protective coatings limit carbonation, while low-strength concrete in dry environments is more vulnerable to deeper penetration.
- Carbonation also contributes to CO₂ sequestration; e.g., the Itaipu Dam absorbed almost 13,384 tons of CO₂ in 35 years, showing potential for passive carbon capture in concrete infrastructure.

8 Future Recommendations

- Develop cost-effective, advanced sensors and testing methods for real-time monitoring of concrete carbonation under varied conditions.
- Explore innovative restoration and repair techniques to counteract carbonation-induced degradation in concrete structures.
- Investigate anti-carbonation strategies, including surface coatings, densifiers, and hybrid cementitious systems, to protect reinforced concrete.
- Conduct long-term field validation studies to improve carbonation models and service-life predictions, considering CO₂ exposure, humidity, and pore structure changes.

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

Funding: This research received no specific grant from any funding agency in the public, commercial, or non-profit sectors.

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