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Accelerated and natural carbonation of high-volume limestone powder concrete: an experimental study

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ABSTRACT

Carbonation-induced corrosion is a primary durability concern for reinforced concrete structures in continental climates. At the same time, efforts to mitigate the environmental impact of cement production have driven the development of low-clinker concrete, including those incorporating high volumes of limestone powder (HVLPC). Although these concretes offer significant sustainability benefits, their carbonation resistance, often compromised due to lower alkaline reserve, remains a critical challenge. In addition to a reference mixture, nine HVLPC mixtures were produced with cement mass replacement levels of 30%, 45%, and 55%, using three limestone powders of different fineness. All mixtures were designed to achieve similar workability and compressive strength. Carbonation was monitored under accelerated conditions for 56 days and under natural laboratory exposure for two years. The results indicate a pronounced reduction in carbonation resistance with increasing limestone powder content. While moderate replacement levels (up to 30%) caused relatively limited changes, mixtures with higher substitution levels (≥ 45%) led to a pronounced increase in carbonation depth. The effect of particle size was found to be secondary and inconsistent. Analysis confirmed that while the square-root-of-time relationship adequately describes carbonation kinetics under accelerated conditions, even for replacement levels exceeding 50%, direct extrapolation to natural exposure remains unreliable. Findings indicate that existing models correlating accelerated and natural carbonation depths are not directly applicable to HVLPC. To overcome this limitation, a modification of the *fib* Model Code 2010 was proposed by introducing empirical coefficients for different replacement ranges (0–30% and 45–55%). This modification ensured a more accurate and reliable prediction.

1 Introduction

It has been reported that carbonation is responsible for a significant proportion of damage in reinforced concrete structures, especially in urban, non-marine environments [1–3]. Some studies suggest that a large portion, around 65-70%, of reinforced concrete structures are at least partially affected by carbonation during their service life, although the extent strongly depends on environmental exposure and material properties [4–7]. Despite its importance, carbonation was historically underestimated compared to other deterioration mechanisms, particularly in earlier design practices that assumed sufficient durability through high cement content and low permeability. However, the traditionally high cement content used to mitigate durability issues is increasingly challenged due to its substantial environmental impact, with cement production contributing approximately 7–10% of global anthropogenic CO_2 emissions [8,9].

Carbonation is a physicochemical process in which atmospheric CO_2 penetrates the concrete pore system and reacts with calcium hydroxide ($Ca(OH)_2$) and other hydration products, leading to a reduction in pore solution alkalinity. Once the pH drops below a critical threshold (9–10), the passivation layer that protect embedded steel reinforcement is destabilized, making reinforcement susceptible to corrosion [10].

In the contemporary context of sustainable construction, the increased use of supplementary cementitious materials (SCM) and fillers, while beneficial in reducing CO_2 emissions, may have unintended consequences on durability performance. In particular, concretes with reduced clinker content generally exhibit lower calcium hydroxide content and altered pore structure, which can accelerate carbonation rates [11,12]. This has raised concerns

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regarding the long-term performance of these concretes, especially in environments where carbonation is the dominant deterioration mechanism.

Over the past two decades, extensive research has been devoted to understanding the carbonation behavior of concrete containing SCMs and mineral fillers. It is well established that the incorporation of SCMs generally alters both the chemical buffering capacity and the pore structure of concrete, which are the two key parameters governing carbonation resistance [13,14]. While some pozzolanic materials tend to refine the pore structure over time, their lower calcium hydroxide content often results in increased carbonation depth, particularly at early ages [11,15]. However, the influence of limestone powder on carbonation resistance remains a subject of ongoing research and debate.

Several studies have reported an increased carbonation depth in concretes with high limestone content, primarily due to the dilution of clinker and reduced alkalinity [14,16–18]. On the other hand, improvements in microstructure due to filler effects and optimized particle packing may partially mitigate this negative impact, especially at lower replacement levels [19]. Bertolini et al. [20,21] also obtained different results in their works, especially at lower replacement percentages (up to 15%). The effect becomes even more complex in the case of high-volume limestone powder concrete (> 35%), where cement replacement levels significantly exceed those traditionally allowed in standards. However, at such high replacement levels, a distinctly negative trend is observed regarding the addition of limestone powder and its effect on the carbonation resistance of concrete [22–24].

Years of research have resulted in numerous recommendations and the development of new or by modifying existing models for predicting the carbonation resistance of concrete, incorporating different types of supplementary cementitious materials [25–27]. Meanwhile, Radović et al. [24] are refining the existing *fib* model [28], originally formulated for OPC concrete, extending its applicability to concretes with a high limestone powder content. Based on this modification, they proposed adjusted (higher) values for concrete cover depth compared to those prescribed for conventional OPC concretes [29]. These revised recommendations take into account the specificities related to the carbonation resistance of high-volume limestone powder concrete (HVLPC) and aim to ensure the required service life of 50 and 100 years.

2 Objectives

Despite the significant research, most available studies rely predominantly on accelerated carbonation testing, typically conducted under elevated CO_2 concentrations. While these methods enable rapid comparison between mixtures, their ability to accurately represent natural carbonation processes remains limited. Consequently, there is still a lack of comprehensive experimental studies that combine both accelerated and long-term natural carbonation data for HVLPC. This gap is especially critical in the context of modern sustainable concrete design, where high levels of clinker substitution are increasingly being considered. In this context, the present study aims to investigate the carbonation resistance of high-volume limestone powder concrete through a comprehensive experimental program. Both accelerated and natural carbonation exposures are considered in order to evaluate the influence of limestone powder content and fineness on carbonation kinetics.

3 Experimental research

3.1 Component materials and concrete mixture proportions

All concrete mixtures were produced using Ordinary Portland cement CEM I 42.5R in accordance with EN 197–1 [30], with the median particle size of approximately d_{50} = 11.1 µm and a particles density of about 3100 kg/m^3.

To investigate the influence of limestone powder fineness, three limestone powders with different particle size distributions were employed: L1 (d_{50} = 2.9 µm), L2 (d_{50} = 5.4 µm), and L3 (d_{50} = 11.7 µm). These limestones contain approximately 98% $CaCO_3$ [31] and meet the requirements of the EN 197-1 [30] in terms of chemical composition. The density of limestone powder particles was in the range of 2690±30 kg/m^3. Particle size distributions with the corresponding curves are shown in Figure 1.

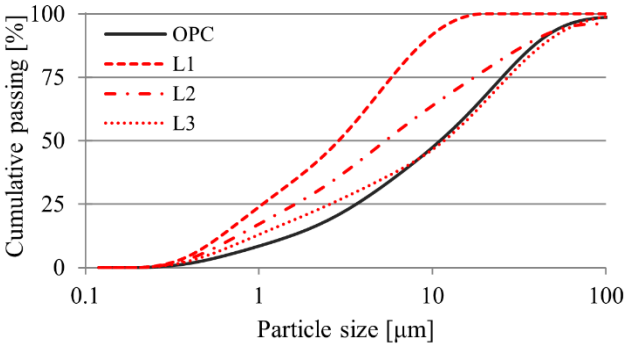

Figure 1. Particle size distribution of cement and limestone powders

Natural river aggregate originating from the Danube River was used, consisting of three fractions: 0–4 mm, 4–8 mm, and 8–16 mm. The oven-dried density of the aggregate grains varied in the range of 2594-2660 kg/m^3. After drying the aggregate, its particle size distribution was determined by dry sieving in accordance with EN 933-1 [32]. The grading curves of the tested aggregate fractions are presented in Figure 2. In the same figure, the reference curves, indicated by dashed lines, represent the limiting envelopes for the respective fractions SRPS B.B3.100 [33] and SRPS B.B2.01 [34]. The grading curves for all aggregate fractions showed slight deviations from the recommended ranges. Fraction I exhibited a somewhat lower amount of fine particles, with cumulative passing of approximately 5% on the 0.25 mm sieve and 1% on the 0.125 mm sieve, compared to the recommended values of 8% and 2%, respectively. Fraction II contained approximately 19% of undersized particles passing the 4 mm sieve, exceeding the recommended 15%. Fraction III exhibits a marginally increased proportion of oversized particles, around 2% retained on the 16 mm sieve and the intermediate 11.2 mm sieve. It can be concluded that these deviations are minor and do not hinder the practical use of the aggregate. It should also be noted that compliance with the reference grading envelopes is not strictly mandatory, as these limits are primarily intended as guidelines rather than strict acceptance criteria.

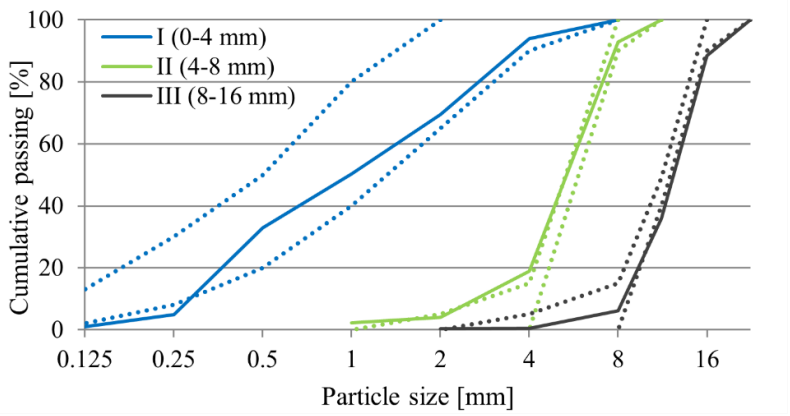

Figure 2. Particle size distribution of natural aggregate

The proportioning of individual aggregate fractions in the mixture was determined using the Funk–Dinger model [35,36], which was originally developed based on computer simulations assuming ideally spherical particles. The model can be expressed as follows:

$$Y_{(d)} = \frac{d^n - d_{min}^n}{d_{max}^n - d_{min}^n} \quad (1)$$

where:

$Y_{(d)}$ – Cumulative passing through a sieve with an opening d

d_{max} – nominal value of the maximum particle size [mm]

d_{min} – nominal value of the minimum particle size [mm]

n – exponent that governs the distribution between finer and coarser particles, i.e., it controls the packing density of the mixture

One limitation of this approach is that it does not consider the actual particle shape, which may significantly influence particle packing density. Furthermore, it can be questioned whether such a simple formulation can adequately describe a system in which particle sizes differ by several orders of magnitude (up to 10^4–10^5 times). Nevertheless, for the commonly adopted exponent value of $n = 0.37$, the model yields the maximum packing density, which has been confirmed by numerous experimental studies [35,37–39].

The recommended range of aggregate particle size distribution (curves A, B, and C) is defined by the SRPS U.M1.206 [40]. Figure 3 presents these reference curves, which delineate the optimal grading envelope for concrete, along with the Funk–Dinger curves corresponding to exponent values of 0.37 and 0.32. A lower exponent value results in a higher proportion of fine particles, which generally improves workability, although it may lead to a slight reduction in compressive strength.

It can be observed that the grading curves obtained using the Funk–Dinger model lie within the reference envelope bounded by curves A and C over nearly the entire particle size range from 0.25 to 16 mm. In practice, it is not necessary for the particle size distribution of a concrete mixture to exactly match a specific reference curve or to remain entirely within the prescribed grading envelope. If experimental results confirm that the designed mixture meets the required performance criteria, minor deviations from the recommended ranges can be considered acceptable [41].

The adopted total amount of aggregate was approximately 1850 kg/m3 (±1%) in all mixtures, and the shares of each fraction were 52%, 21%, and 27%, respectively. The particle size distribution curve of the OPC mixture, with minor local

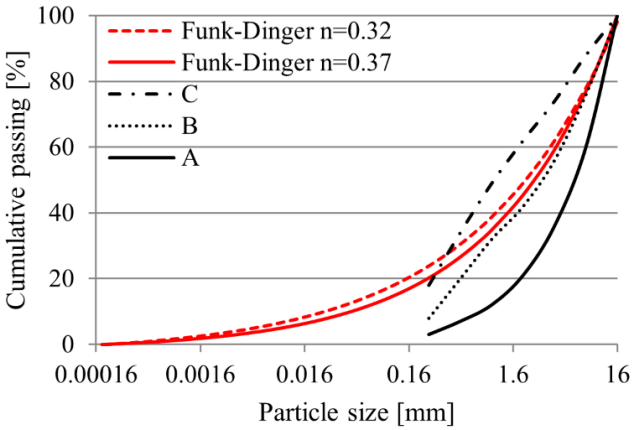

Figure 3. Comparison of different models for selecting aggregate particle size distribution

deviations, closely follows the region bounded by the Funk–Dinger curves corresponding to exponent values of 0.37 and 0.32. A similar trend can be observed for the other mixtures; however, due to the presence of limestone powder, these contain a higher proportion of very fine particles (< 0.1 mm), which results in more noticeable deviations in this size range.

A polycarboxylate-based superplasticizer was used to regulate the workability of fresh mixtures, with a target slump of at least 200 mm (consistency classes S4–S5 according to EN 206-1 [30]).

Final concrete mix compositions were determined using the absolute volume method. The reference mixture (OPC) was prepared with 330 kg/m³ of cement and a water-to-cement ratio (*w/c*) of 0.51. Cement was partially replaced by mass with 30%, 45% and 55% of limestone powders L1, L2 and L3, while *w/c* ratios ranged between 0.62 and 0.75. In mixtures containing limestone powder, lower effective *w/c* ratios were required to maintain the desired strength level and target compressive strength (approximately $f_{cm,cube}$ = 47 ± 6 MPa on 150 mm cubes) corresponding to commonly used structural concrete classes C25/30 and C30/37 [42]. This led to a reduction in cement paste volume, which was compensated by increasing the amount of limestone powder beyond the mass of the replaced cement. In this way, the overall particle packing and rheological properties of the mixtures were improved. The detailed mixture proportions and designations, defined by the type of limestone powder and percentage of cement replacement, are summarized in Table 1.

Table 1. Proportions of paste constituents in concrete mixtures

Concrete mix	Cement [kg/m^3]	Limestone [kg/m^3]	Water [kg/m^3]	w/c [–]	Superplast. [%]
OPC	330	0	169	0.51	1.0
L1-30%	230	200	143	0.62	1.5
L2-30%	230	200	143	0.62	1.5
L3-30%	230	200	143	0.62	1.5
L1-45%	180	250	126	0.70	2.0
L2-45%	180	250	126	0.70	2.0
L3-45%	180	250	127	0.70	2.0
L1-55%	150	280	112	0.75	2.0
L2-55%	150	280	112	0.75	2.0
L3-55%	150	280	112	0.75	2.0

3.2 Preparation, casting, curing, and testing of concrete

Each concrete mixture was mixed for a total duration of approximately five minutes. The consistency of fresh concrete was verified using the standard slump test in accordance with EN 12350-2 [43]. Immediately after testing, the mixtures were placed into moulds and compacted using vibrating table without delay. After removal from the moulds, all samples were subsequently cured in water at a temperature of 20 ± 2°C for 28 days. Compressive strength was determined at the age of 28 days on 150 mm cube specimens, following EN 12390-3 [44]. For each mixture, three specimens were tested and the average value was reported.

The resistance to carbonation was evaluated using prismatic specimens with dimensions of 120×120×360 mm, with two prisms prepared per mixture. After the initial 28-day curing period, all specimens were subjected to an additional conditioning phase lasting 14 days in a controlled climate chamber (20 ± 2°C and 65 ± 5% relative humidity) to achieve a uniform internal moisture state prior to CO_2 exposure. Following conditioning, each prism was split into two parts. One half was used for accelerated carbonation testing, while the other was reserved for natural carbonation exposure. Accelerated carbonation (ACC) was performed in a chamber under controlled conditions (2% CO_2 concentration, temperature of 20 ± 2°C, and relative humidity of 65 ± 5%), in line with *fib* recommendations [28,45]. The duration of exposure was 56 days, with intermediate measurements conducted after 7, 14, 21, 28, and 56 days.

Specimens designated for natural carbonation (NAC) were stored under ambient laboratory conditions for a period of 2 years. Environmental parameters that were continuously measured during this period included temperature, relative humidity, and CO_2 concentration and are shown, along with their mean values, in Figure 4.

Carbonation depth was determined using the phenolphthalein indicator method, in accordance with EN 14630 [46], Figure 5. For each specimen, measurements were taken at eight points on each exposed surface, and the reported values represent the mean carbonation depth for both accelerated and natural exposure conditions.

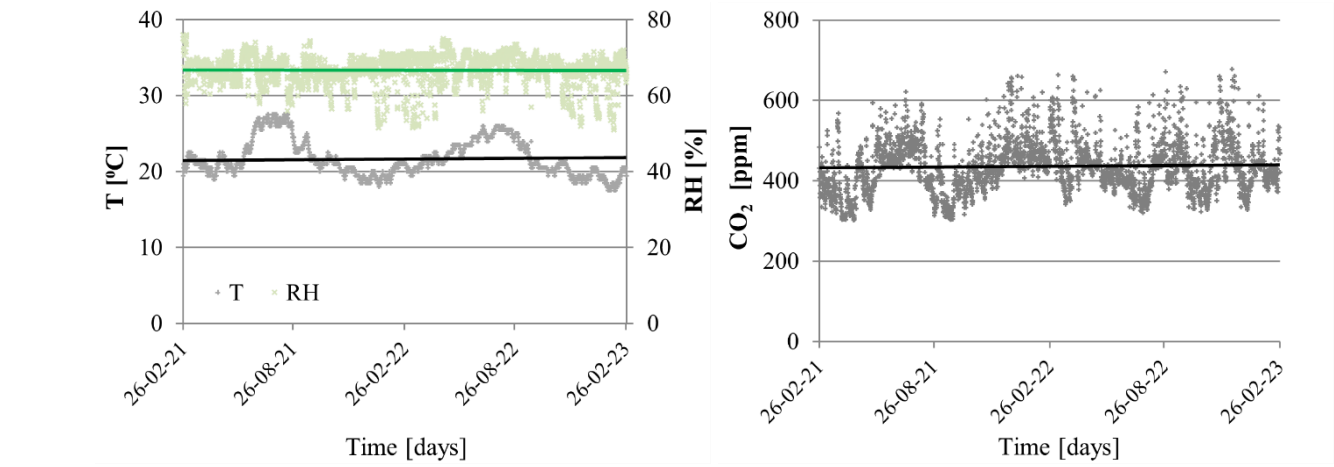

Figure 4. Environmental parameters during natural carbonation test

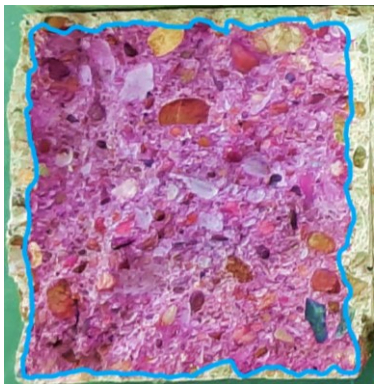

Figure 5. Appearance of a sample prepared for measuring carbonation depth after surface treatment with phenolphthalein solution

4 Results and discussion

4.1 Workability and compressive strength

All concrete mixtures achieved a high level of workability, corresponding to consistency classes S4 and S5 [47], with slump values between 200 and 250 mm, Figure 6. Such consistency indicates excellent flowability and confirms that all mixtures are well suited for applications such as pumped concrete. It was observed that mixtures with higher levels of cement replacement required increased dosages of superplasticizer to maintain the same workability. This behavior can be explained by the reduced absolute water content in these mixtures. Although the nominal *w/c* ratios were higher compared to the reference OPC mixture, the lower cement content resulted in a reduced volume of free water available for lubrication of the mix, thereby necessitating additional chemical admixture. It should also be noted that, if slightly lower workability is acceptable, the dosage of superplasticizer could be reduced. This would have a direct positive impact on the overall cost-effectiveness of the mixtures [48].

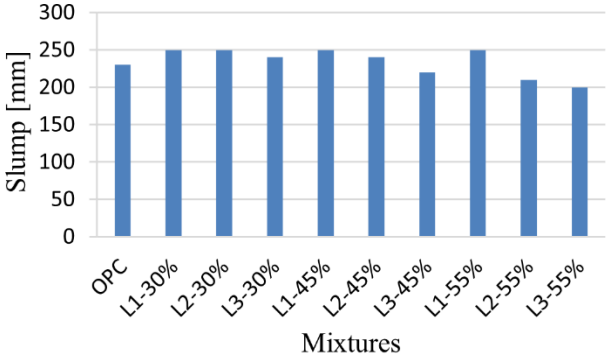

Figure 6. Measured slump values

The compressive strength of the tested concrete mixtures at the age of 28 days, determined on cube specimens, is presented in Figure 7. It can be observed that all mixtures satisfied the target strength requirements corresponding to concrete classes C25/30 and C30/37, with measured values ranging between 41 and 53 MPa. The highest compressive strength was recorded for mixture L1-30%, which exceeded the reference mixture by approximately 8%. This improvement can be attributed to the filler and packing effects of finely ground limestone powder. The presence of very fine particles enhances particle packing, reduces voids within the matrix, and leads to a denser microstructure, which positively affects strength development.

Mixtures L2-30% and L1-45% followed, exhibiting compressive strength values very close to OPC concrete. In contrast, the remaining mixtures generally showed slightly lower strengths, with most values being 1.5–8% below the OPC. The lowest compressive strengths were observed for mixtures L3-45% and L3-55%, which exhibited reductions of approximately 17% and 15%, respectively, compared to the OPC mixture. The coarser particle size of the limestone powder (L3) results in a less pronounced packing effect, which contributes to a decrease in strength. Nevertheless, all mixtures remained within the targeted strength range, confirming their suitability for structural applications.

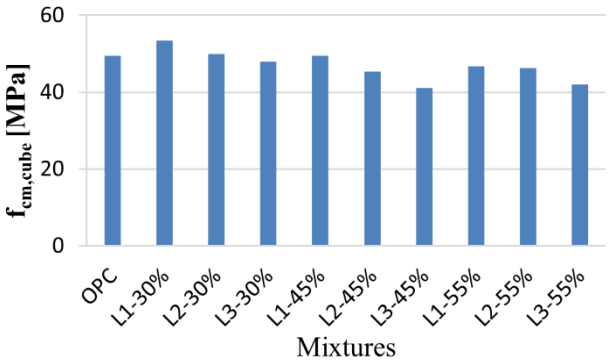

Figure 7. Compressive strength of concrete mixtures at 28 days

4.2 Accelerated and natural carbonation

The carbonation depths obtained from the accelerated test after 28 and 56 days of exposure are presented in Figure 8. A clear negative effect of increasing limestone powder content on the carbonation resistance of concrete can be observed. Mixtures with 30% cement replacement exhibited carbonation depths that were 9–28% higher than that OPC concrete after 28 days of exposure. With further increases in limestone powder content beyond 30%, the reduction of carbonation resistance becomes more pronounced, accompanied by greater scatter in the results. During the same exposure period, mixtures with 45% cement replacement showed carbonation depths that were 90–157% higher than the OPC mixture. Comparable values were also observed for mixtures with 55% cement replacement.

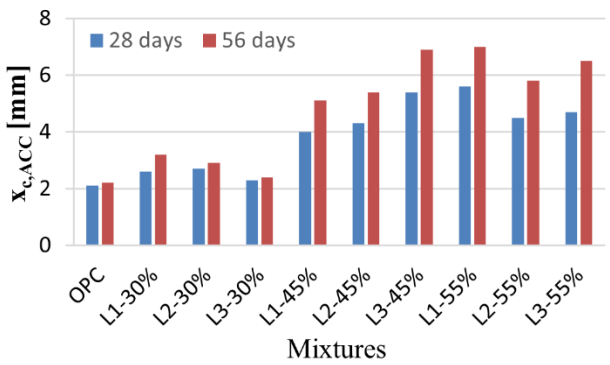

Figure 8. Measured carbonation depths after 28 and 56 days of exposure

Extending the exposure period to 56 days led to only a modest increase (approximately 5%) in carbonation depth for the reference concrete, whereas mixtures containing limestone powder exhibited significantly larger increases, ranging from 13% to 40%, depending on the replacement level. This indicates that the differences in carbonation resistance between conventional OPC concrete and HVLPC become more pronounced with prolonged exposure.

Based on the presented results, the influence of limestone powder fineness on accelerated carbonation resistance cannot be clearly established. For mixtures with 30% cement replacement, those incorporating the coarser powder (L3) performed slightly better, while at 45% replacement the best results were achieved with the finest powder (L1), and at 55% replacement with the intermediate powder (L2). Overall, these findings suggest that the particle size distribution of limestone powder does not have a decisive influence on carbonation resistance under accelerated conditions.

According to the square-root theory, the carbonation depth can be related to the exposure time in accordance with Fick's second law, as expressed by the following relationship (Tuutti, 1982):

$$x_c = k_c \cdot t^{0.5} \tag{2}$$

For given environmental conditions (e.g., CO_2 concentration, relative humidity, and temperature), a linear relationship is typically observed between the carbonation depth (x_c) and the square root of exposure time ($t^{0.5}$). This relationship is characterized by the carbonation coefficient (k_c), which serves as a useful parameter for comparing the carbonation resistance of different concrete mixtures. The same formulation has been adopted in the latest version of EN 12390-10 [49].

Figure 9 illustrates the evolution of carbonation depth under accelerated conditions as a function of exposure time (7, 14, 21, 28 and 56 days), together with the corresponding values of the carbonation coefficient obtained through regression analysis. Even under accelerated carbonation conditions, a strong linear correlation between carbonation depth and the square root of time is observed. This is confirmed by the very high coefficients of determination (R^2 = 0.97–1.00) obtained for all mixtures, indicating that the square-root model provides an excellent representation of the carbonation process within the investigated time frame. These observations confirm that the square-root model is still fully applicable even at very high limestone powder contents.

The obtained carbonation coefficients show a clear and consistent increase with the incorporation of limestone powder. The reference OPC mixture exhibited the lowest value, approximately 0.35 mm/day$^{0.5}$. At 30% cement replacement, the k_c values increase gradually to 0.39–0.46 mm/day$^{0.5}$, corresponding to a 11–31% increase compared to the OPC concrete. Within this group, the best performance was achieved with the coarsest limestone powder (L3), while the finest powder (L1) resulted in the highest carbonation rate. This indicates that, at moderate replacement levels, the beneficial effect of improved packing is not sufficient to compensate for the reduced alkaline reserve.

At 45% replacement, carbonation resistance is further reduced, these values increase significantly to about 0.75 mm/day$^{0.5}$ (L1-45%), 0.79 mm/day$^{0.5}$ (L2-45%), and 0.98 mm/day$^{0.5}$ (L3-45%), corresponding to an increase of about 114–180% compared to OPC concrete. In this case, the influence of particle size becomes less systematic, suggesting that the dominant factor is no longer packing density but the reduced cement content and increased porosity associated with high cement substitution. These results indicate a transitional behavior where dilution effects begin to govern carbonation performance.

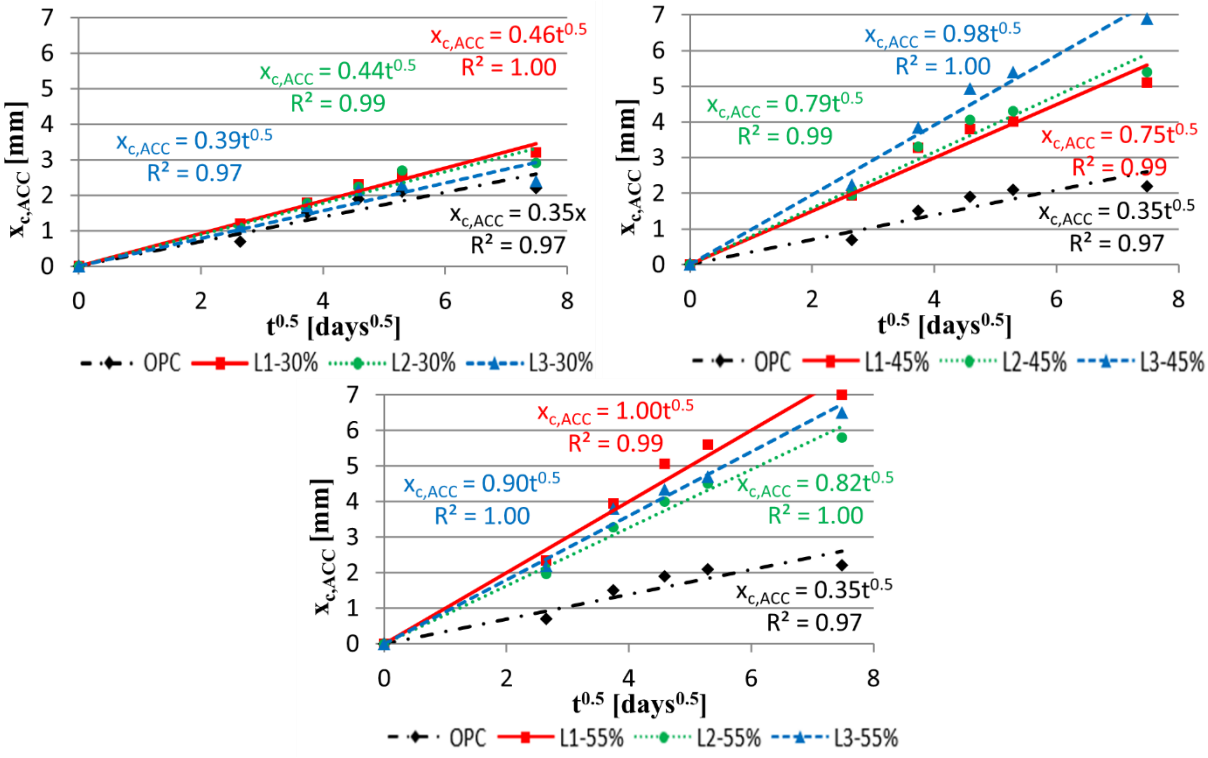

Figure 9. Development of accelerated carbonation over time for mixtures with different cement replacements

Finally, increased cement replacement level up to 55% does not lead to a drastic further increase in the carbonation coefficient. The k_c values remain within almost identical limits as the previous ones 0.82–1.0 mm/day$^{0.5}$, with a minor change. Within this group, mixtures with coarser limestone powder again show slightly better resistance, while the finest powder exhibits the highest carbonation rate. Overall, the results confirm that high levels of cement replacement significantly impair carbonation resistance, with particle size effects becoming secondary to the dominant influence of cement dilution.

This behavior can primarily be attributed to the reduced alkaline reserve of such systems [14], which lowers their buffering capacity against CO_2 ingress. In addition, the dilution effect, resulting from reduced clinker content, leads to a lower amount of hydration products, while changes in pore structure and potential increases in porosity associated with calcium carbonate formation [50] further contribute to the reduced carbonation resistance. Although finer particles may improve packing density, their beneficial effect is insufficient to counterbalance these dominant mechanisms at high replacement levels.

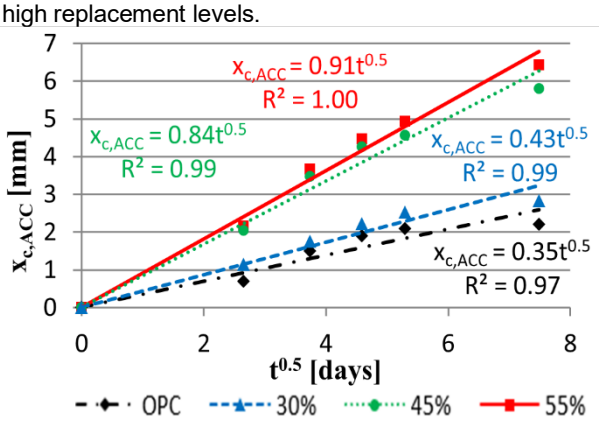

Figure 10. Development of accelerated carbonation over time - averaged for different cement replacement percentages

A comparison of the average carbonation coefficients across all mixture groups further confirms the dominant role of cement replacement level, Figure 10. While the increase at 30% remains moderate (≈ 23%), a sharp increase occurs between 30% and 45%, where the carbonation rate nearly doubles. These results suggest the existence of a critical threshold at approximately 40–45% cement replacement, beyond which carbonation resistance deteriorates rapidly. Below this level, the negative effects of clinker reduction can still be partially mitigated through improved particle packing and microstructural densification. Although limestone powder fineness influences results within individual groups, its overall effect is secondary compared to the replacement level. However, at higher replacement levels, the loss of alkaline reserve becomes the governing factor controlling carbonation behavior.

Figure 11 presents the measured carbonation depths under natural exposure conditions after two years. A comparison of the results clearly indicates significantly higher carbonation depths in mixtures containing limestone powder compared to the reference OPC concrete. Among all mixtures, L1-30% showed the smallest deviation, with a carbonation depth approximately 8% higher than that of the reference. Mixtures L2-30%, L3-30%, and L1-45% exhibited increases of around 25%. In contrast, the remaining mixtures showed substantially higher carbonation depths, ranging from 67% to 82% above the OPC mixture, despite relatively modest reductions in compressive strength (only 6–15%). These findings highlight that, even when comparable compressive strength is achieved, concrete with limestone powder remains more susceptible to carbonation.

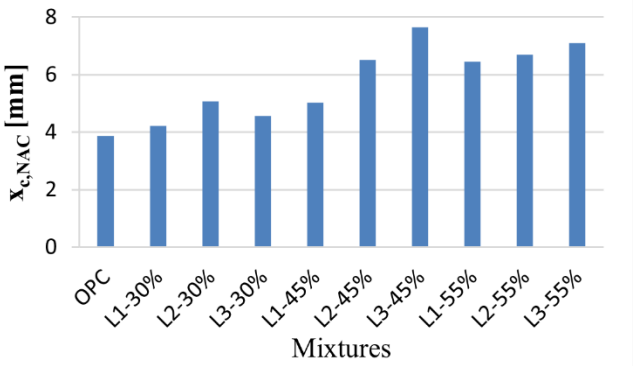

Figure 11. Measured carbonation depths after 2 years

It is also noteworthy that the differences in carbonation depth between the OPC and limestone-containing mixtures were more pronounced in the accelerated tests. This may indicate that carbonation kinetics in limestone powder concrete differ under elevated CO_2 concentrations, suggesting that accelerated testing conditions may amplify these differences.

Unlike the accelerated tests, the influence of limestone powder fineness is more evident under natural exposure conditions. Mixtures incorporating the coarser powder (L3) exhibited carbonation depths that were 10% to 54% higher than those prepared with the finest powder (L1). Similar trends have been reported by other researchers [51,52], confirming the beneficial role of finer particles in improving the microstructure and, consequently, the resistance to carbonation. However, contradictory results have also been documented in the literature, even under natural carbonation conditions [23].

Measured carbonation depths under accelerated conditions (56 days) and natural exposure (2 years), together with their relationships with *w/c* ratio and cement content, are presented in Figure 12 (left and right, respectively). Carbonation depth increases significantly with increasing *w/c* ratio and decreasing cement content under both exposure conditions. In both cases, this effect is more pronounced for accelerated carbonation. These trends reflect the combined influence of increased porosity at higher *w/c* ratios and reduced alkaline reserve at lower cement contents, noting that mixtures with lower cement content also had higher *w/c* ratios.

The steeper trends observed under accelerated conditions indicate differences in carbonation kinetics, suggesting that accelerated testing amplifies the sensitivity of carbonation depth to mixture parameters. Moreover, the differing slopes of the curves confirm that the relationship between natural and accelerated carbonation is not uniform across all mixtures. The largest discrepancy between natural and accelerated carbonation was observed for the OPC mixture (approximately 76%), while this difference gradually decreased with increasing *w/c* ratio (i.e., decreasing cement content). For mixtures with the highest *w/c* ratio, carbonation depths under natural and accelerated conditions became nearly equal (average difference of about 5%), with the curves almost intersecting at this point. Finally, slightly higher coefficients of determination for accelerated carbonation (R^2 = 0.81–0.84) compared to natural conditions (R^2 = 0.68–0.69)

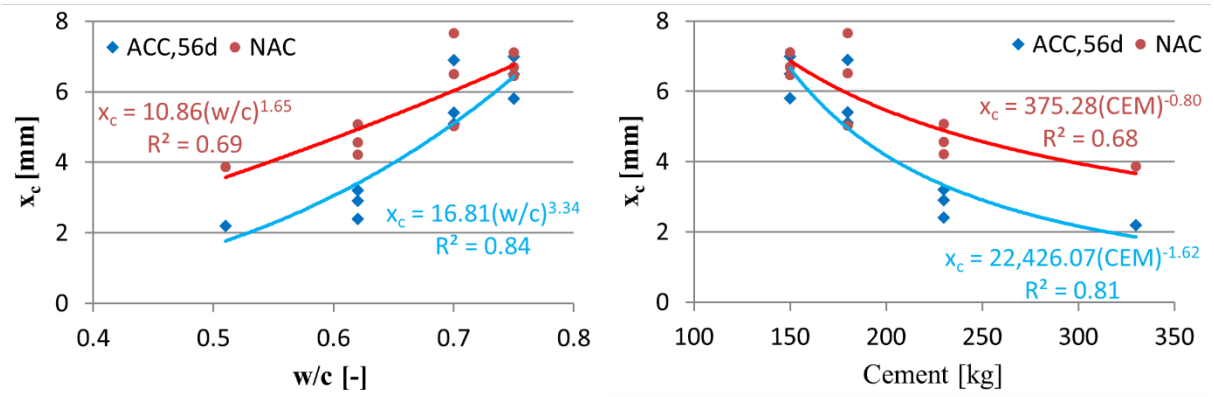

Figure 12. Accelerated (56 days) and natural (2 years) carbonation regarding to water-to-cement ratio (left) and cement content (right)

indicate a more consistent relationship between mixture parameters and carbonation depth under accelerated testing. The cause of this phenomenon may be an increase in the internal humidity of the concrete due to water generated in the carbonation reaction [53,54]. The higher the CO_2 concentration, the greater the amount of water produced, which slows down the process. At low CO_2 concentrations, the internal humidity cannot reach a level higher than the external humidity, so the process is unlikely to change.

5 Prediction of carbonation depth based on accelerated tests and exposure time

The carbonation depth relationship, Eq. (2), can be expressed in terms of carbon-dioxide concentration as:

$$x_c = K \cdot \sqrt{CO_2} \cdot t^{0.5} \tag{3}$$

where the coefficient *K* depends on concrete properties, relative humidity, and ambient temperature, but is independent of CO_2 concentration, which appears as an explicit variable in the equation.

Standards for carbonation depth assessment specify different reference CO_2 concentrations. To compare carbonation depths at different conditions, the following relationship can be derived for the same concrete type [15,27]:

$$\frac{x_{c,1}}{x_{c,2}} = \sqrt{\frac{|CO_2|_1}{|CO_2|_2}} \cdot \left(\frac{t_1}{t_2}\right)^{0.5} \tag{4}$$

The determination of carbonation coefficients under natural exposure conditions typically requires several years, which makes this approach impractical for direct use in structural design. For this reason, accelerated carbonation testing is widely used, reducing the testing period from years to days or weeks. However, carbonation depths under natural conditions remain the primary input for durability design of reinforced concrete structures, as they directly govern concrete cover requirements and service life predictions.

Since the environmental conditions in this research are comparable between natural and accelerated tests, and the coefficient K is independent of CO_2 concentration, the carbonation depth under natural exposure can be estimated from accelerated test results using:

$$x_{c,NAC}(t) = x_{c,ACC} \sqrt{\frac{|CO_2|_{NAC}}{|CO_2|_{ACC}}} \cdot \left(\frac{t_{NAC}}{t_{ACC}}\right)^{n} \tag{5}$$

where n is the time exponent that accounts for differences in carbonation kinetics between natural and accelerated exposure conditions.

A commonly adopted value of the time exponent is *n = 0.5*, consistent with the classical square-root-of-time relationship for carbonation depth [1]. However, alternative values have been proposed to account for deviations from ideal diffusion-controlled behavior. For example, *n = 0.4* was suggested by Sisomphon and Franke [15] to incorporate pore refinement effects during carbonation, while Carević et al. [27] reported *n = 0.78* for concretes with high fly ash content.

Based on the previously defined relationship, the value of exponent can be determined for known quantities as:

$$n = \frac{log\left(\frac{\frac{x_{c,NAC}}{x_{c,ACC}}}{\sqrt{\frac{|CO_2|_{NAC}}{|CO_2|_{ACC}}}}\right)}{log\left(\frac{t_{NAC}}{t_{ACC}}\right)} = \frac{log\left(\frac{x_{c,NAC}}{x_{c,ACC}} \cdot \sqrt{\frac{|CO_2|_{ACC}}{|CO_2|_{NAC}}}\right)}{log\left(\frac{t_{NAC}}{t_{ACC}}\right)} \tag{6}$$

Using this expression and the experimental results, the exponent *n* was evaluated for each mixture and exposure period. The obtained values, summarized in Figure 13, show

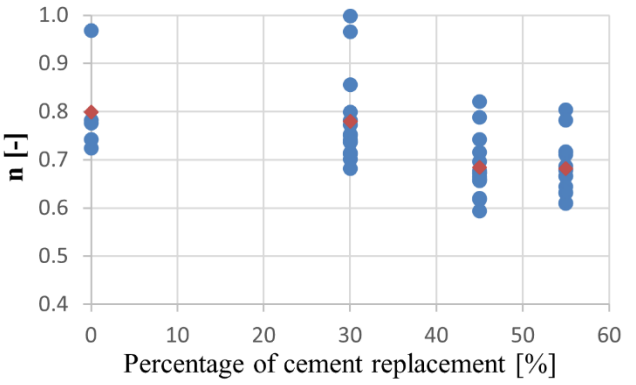

Figure 13. Exponent values as a function of cement replacement percentage with mean values for each group

notable scatter within individual mixture groups. Nevertheless, the average values reveal a clear trend: OPC and mixtures with 30% cement replacement exhibit similar behavior (n = *0.78*), while mixtures with 45% and 55% replacement yield lower values (n = *0.68*).

This observation supports grouping the mixtures into two categories: (I) 0–30% and (II) 45–55% cement replacement. To preserve the classical square-root formulation (n = *0.5*) and avoid introducing a variable exponent, the data were reformulated by analyzing the relationship between $\frac{x_{c,NAC}}{x_{c,ACC}\cdot\sqrt{|CO_2|_{NAC}/|CO_2|_{ACC}}}$ and $\frac{t_{NAC}}{t_{ACC}}$ for both groups (Figure 14). The high coefficients of determination (R^2 = 0.80 and 0.89) confirm the validity of this approach.

Based on these results, modified expressions for carbonation depth under natural conditions are proposed:

- For 0–30% replacement:

$$x_{c,NAC}(t) = 2.65 \cdot x_{c,ACC}(t)\sqrt{\frac{|CO_2|_{NAC}}{|CO_2|_{ACC}}}\cdot\left(\frac{t_{NAC}}{t_{ACC}}\right)^{0.5} \quad (7)$$

- For 45–55% replacement:

$$x_{c,NAC}(t) = 1.85 \cdot x_{c,ACC}(t)\sqrt{\frac{|CO_2|_{NAC}}{|CO_2|_{ACC}}}\cdot\left(\frac{t_{NAC}}{t_{ACC}}\right)^{0.5} \quad (8)$$

The higher coefficients (as well as the higher exponent) obtained for the 0–30% group do not indicate greater carbonation depths, but rather reflect differences in carbonation kinetics between natural and accelerated conditions, as previously discussed. The differences between natural and accelerated carbonation are captured through empirically derived coefficients.

Overall, the results demonstrate that a single conversion factor between accelerated and natural carbonation is not sufficient, and that the effect of cement replacement level must be explicitly accounted for. The proposed modification of the model preserves its physical basis while significantly improving its predictive capability for HVLPC.

Figures 15–19 present the predicted carbonation depths under natural conditions after 2 years, calculated based on accelerated carbonation results obtained after 7, 14, 28, and 56 days of exposure, together with the corresponding measured values for each mixture. The predictions were carried out using the proposed modified expressions (Eq. 7 and Eq. 8) for the groups with 0–30% and 45–55% cement replacement, respectively. For comparison, results obtained using modified values of the time exponent (n = *0.78* for 0–30% and n = *0.68* for 45–55% replacement) as well as the commonly adopted value n = *0.5* according to the square-root-of-time theory are also shown.

A clear trend can be observed from these figures: the classical formulation with n = *0.5* provided the least accurate predictions, systematically underestimating carbonation depths under natural conditions. For mixtures with 45–55% cement replacement, the predicted values were approximately 50% lower than the measured ones ($x_{c,calc}$ / $x_{c,exp}$ = *0.54*), while for the 0–30% group the underestimation is even more pronounced ($x_{c,calc}$ / $x_{c,exp}$ = *0.38*). The corresponding coefficients of variation remained relatively low (13–15%), indicating consistent but biased predictions. The use of modified time exponents (n = *0.78* and n = *0.68*) significantly improved prediction accuracy. The average ratios between predicted and measured values were 1.07 (CoV ≈ 25%) for the 0–30% group and 1.03 (CoV = 17.7%) for the 45–55% group, indicating good agreement, although with increased scatter.

The highest level of accuracy is achieved with the proposed approach that introduces empirical coefficients while preserving the classical square-root formulation (Eq. 7 and Eq. 8). In this case, the predicted and measured values were in excellent agreement, with average ratios of 1.01 and 0.99 for the two groups, respectively. At the same time, the coefficients of variation remained low (13–15%), confirming both the accuracy and robustness of the proposed model.

In the previous paragraph, the accuracy of the three considered approaches was evaluated in general terms. However, their performance shows noticeable deviations depending on the duration of accelerated carbonation exposure. The smallest deviations were obtained with the proposed approach. For mixtures with 0–30% cement replacement, the largest discrepancies were observed for exposure durations of 7 and 56 days, with coefficients of variation of 19.3% and 17.1%, and corresponding ratios $x_{c,calc}$ / $x_{c,exp}$ of 0.92 and 0.85, respectively. For intermediate exposure periods (14 and 28 days), the predictions are more consistent, with ratios in the range of 1.08–1.11 and lower scatter (CoV = 8.3–9.2%). Higher prediction accuracy is observed for mixtures with 45–55% cement replacement. In this case, both the ratios and scatter remain relatively stable across different exposure durations. For 7 and 56 days, the ratios are 0.90 and 0.92, indicating slightly non-conservative

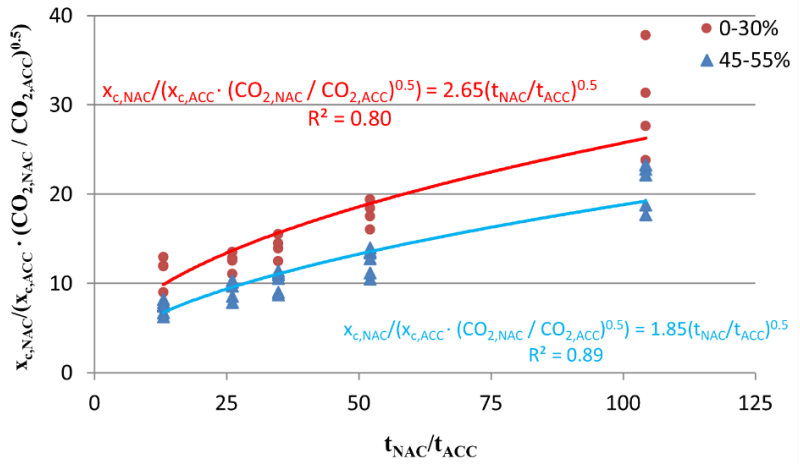

Figure 14. Relationship between $\frac{x_{c,NAC}}{x_{c,ACC}\cdot\sqrt{|CO_2|_{NAC}/|CO_2|_{ACC}}}$ *and* $\frac{t_{NAC}}{t_{ACC}}$ *for 0–30% and 45–55% cement replacement*

estimates, while for intermediate durations the results are generally on the safe side, with ratios between 0.99 and 1.08. The coefficients of variation are consistently low, ranging from 10% to 13%.

These results indicate that the duration of accelerated carbonation exposure has a non-negligible influence on prediction accuracy. Very short exposure periods (e.g., 7 days) may not sufficiently capture the carbonation kinetics, while prolonged exposure (e.g., 56 days) may alter the governing mechanisms due to sustained high CO_2 concentrations. Therefore, an optimal exposure duration should be adopted to ensure reliable correlation with natural carbonation behavior. Overall, the proposed approach shows strong potential for practical implementation in carbonation depth predictions and service life design, particularly by enhancing the reliability of carbonation-based durability predictions and supporting a more rational determination of concrete cover thickness in structures incorporating high volumes of limestone powder.

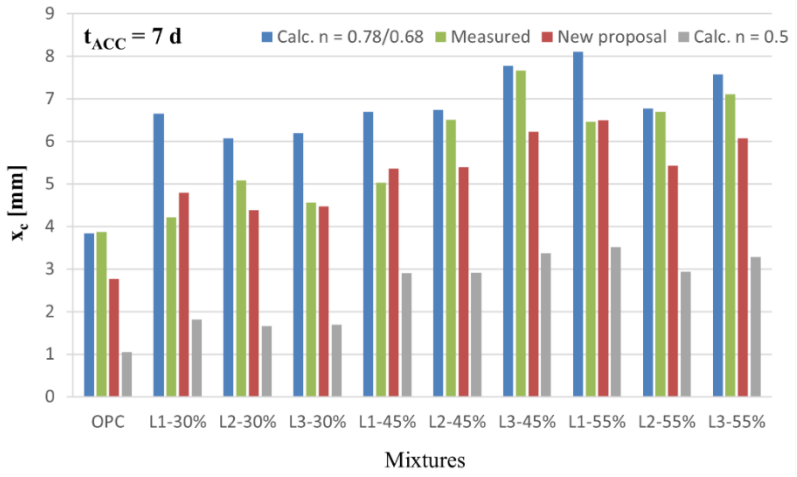

Figure 15. Calculated natural carbonation depths after 2 years based on accelerated carbonation over 7 days

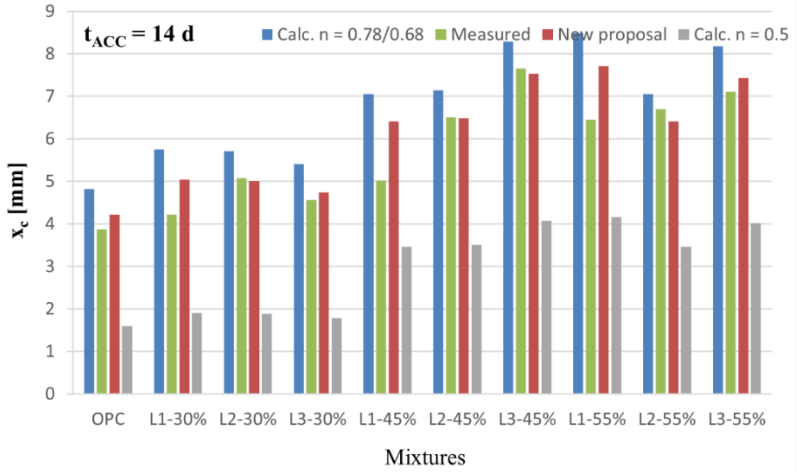

Figure 16. Calculated natural carbonation depths after 2 years based on accelerated carbonation over 14 days

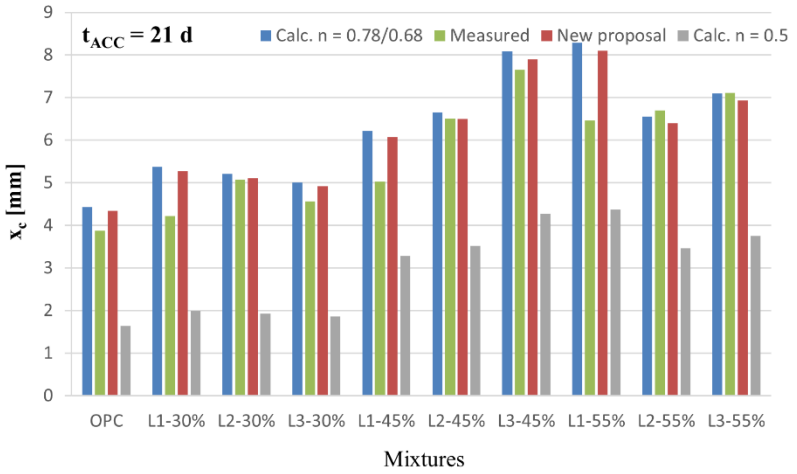

Figure 17. Calculated natural carbonation depths after 2 years based on accelerated carbonation over 21 days

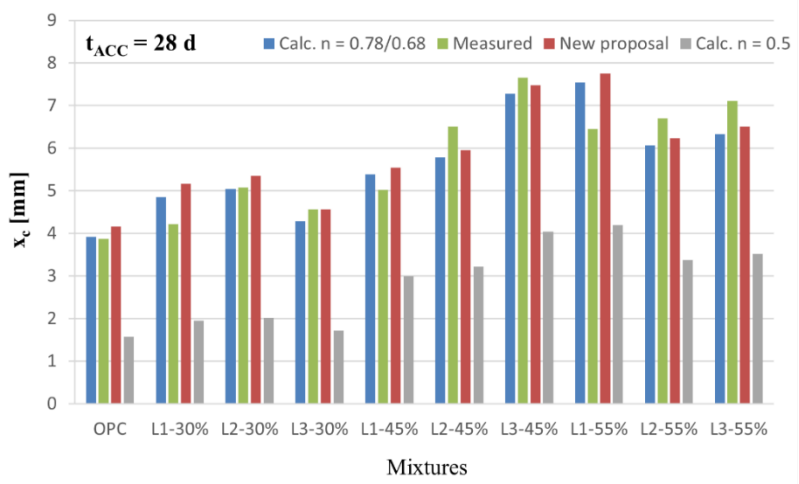

Figure 18. Calculated natural carbonation depths after 2 years based on accelerated carbonation over 28 days

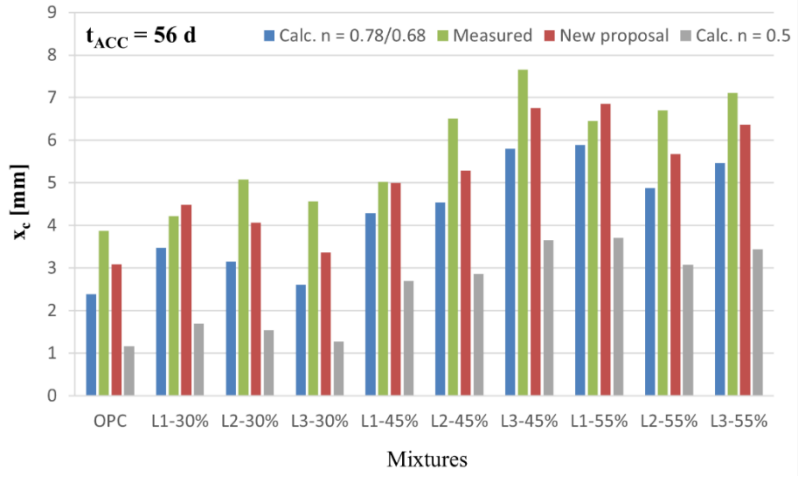

Figure 19. Calculated natural carbonation depths after 2 years based on accelerated carbonation over 56 days

For the sake of clarity, Figure 20 summarizes the ratios between calculated and measured carbonation depths as a function of accelerated exposure duration for all three approaches. Based on the proposed model (Eq. 7 and Eq. 8), the mean values and corresponding coefficients of variation were used to define the 5% and 95% fractiles, forming the confidence interval. Accordingly, no more than 10% of the results are expected to fall outside this range. Using the proposed approach, the predicted carbonation depths deviate from the measured values by up to ±26% for mixtures with 0–30% cement replacement and ±22% for those with 45–55% replacement. Only two data points per group fall outside the confidence bounds. In general, these observations are consistent with the previous discussion, with the largest deviations associated with the shortest (7 days) and longest (56 days) exposure periods.

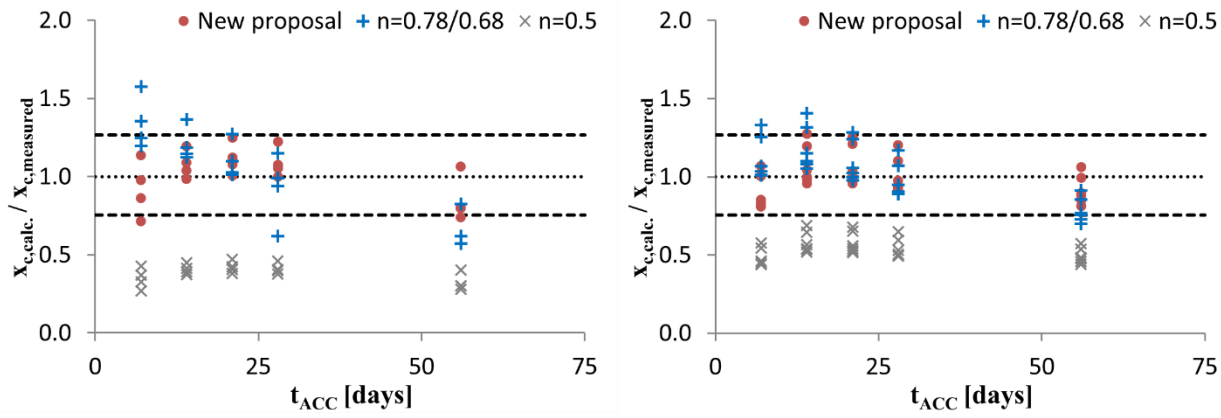

Figure 20. Ratio of calculated and measured carbonation depth for percentage cement replacement 0-30% (left) and 45-55% (right)

The classical formulation with $n = 0.5$ exhibits a similar distribution pattern with respect to exposure duration, but consistently underestimates carbonation depth. In contrast, the approach based on fixed modified exponents ($n = 0.78$ and 0.68) shows the largest scatter and less stable predictions. Together with a noticeable systematic trend with exposure duration, indicating that this formulation introduces a time-dependent bias rather than a stable improvement in prediction accuracy.

6 Conclusions

In this study, an experimental program was conducted to evaluate the influence of limestone powder content and its fineness on the carbonation resistance of concrete. In addition, workability and compressive strength were examined, as these properties are essential for practical structural applications. A total of ten concrete mixtures were produced and tested, including one reference mixture and nine mixtures with cement replaced by limestone powder at levels of 30%, 45%, and 55% by mass. Three limestone powders of the same mineralogical origin but different particle sizes were used.

Based on the experimental results and the performed analysis, the following conclusions can be drawn:

- All mixtures achieved high workability (slump above 200 mm) and met the target compressive strength requirements, with values ranging from 41 to 53 MPa. The highest strength was recorded for mixture L1-30%, exceeding the reference by approximately 8%, while the lowest strengths were observed for L3-45% and L3-55%, with reductions of about 17% and 15%, respectively. The mixtures containing the finest powder exhibiting on average about 6% and 15% higher strengths compared to those with medium and coarse limestone powders.
- Accelerated carbonation results indicate a significant reduction in carbonation resistance with increasing limestone powder content. Mixtures with 30% replacement showed carbonation depths 9–28% higher than the reference after 28 days, while mixtures with 45% and 55% replacement exhibited substantially higher values, in the range of 90–157%. Extending the exposure period from 28 to 56 days had a minor effect on OPC (5% increase), but a much more pronounced effect on HVLPC mixtures (13-40%).
- Development of accelerated carbonation depth over time follows linear trend for all mixtures, indicating that the square-root model remains applicable even at high limestone powder contents. The reference OPC mixture exhibited the lowest k_c value (0.35 mm/day$^{0.5}$), while mixtures with 30% replacement showed moderate increases of 11–31%. At higher replacement levels, a significant rise in k_c was observed, reaching approximately 0.75–0.98 mm/day$^{0.5}$ for 45% and up to 1.00 mm/day$^{0.5}$ for 55% replacement.
- Natural carbonation results after 2 years followed similar trends. The smallest deviation was observed for mixture L1-30% (about 8% higher than OPC), while mixtures L2-30%, L3-30%, and L1-45% showed increases of around 25%. The remaining mixtures exhibited significantly higher carbonation depths, ranging from 67% to 82% above the reference.
- The analysis showed that existing approaches for correlating accelerated and natural carbonation are not directly applicable to HVLPC, significantly underestimating carbonation depth (average calculated-to-measured ratio was 0.47, CoV ≈ 21%). Therefore, a modification of the *fib* Model Code 2010 formulation was proposed. By introducing empirical coefficients (2.65 for 0–30% and 1.85 for 45–55% cement replacement), while retaining the square-root-of-time relationship, a substantial improvement in prediction accuracy was achieved (average calculated-to-measured ratio was 1.00, CoV ≈ 14%).

The presented findings highlight the need for adjusted predictive models when dealing with HVLPC. However, it should be noted that the proposed modification is based only on the experimental results obtained within this study. Keeping this in mind, the conclusions should be interpreted within the limitations of the present study, which considered one cement type (CEM I 42.5R), one aggregate type (river), a specific w/c range (0.51–0.75), accelerated and laboratory natural exposure conditions. Therefore, application to different concrete compositions or in-situ exposure conditions requires additional experimental validation.

Future studies involving: a wider range of materials, concrete strength classes, and environmental conditions (including longer-term natural exposure investigations as well as accelerated carbonation tests under higher CO_2 concentrations) are needed to further assess the robustness and applicability of the proposed approach. Finally, these and similar studies should contribute to the development of broader experimental databases covering a wider range of input parameters. Such databases would enable more reliable calibration and refinement of existing code-based prediction models, thereby supporting the gradual inclusion of HVLPC into future standards provisions.

CRediT authorship contribution statement

A. Radović: Data curation, Investigation, Methodology, Formal analysis, Writing - original draft.

V. Carević: Conceptualization, Methodology, Formal analysis, Writing - review & editing.

A. Savić: Supervision, Resources, Writing - review & editing.

Declaration of competing interest

The authors declare no conflict of interest.

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