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Field assessment of the relationship between natural and accelerated concrete carbonation resistance

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ABSTRACT

The relationship between natural and accelerated concrete carbonation resistance has been investigated. Cores were taken from real structures and their non-carbonated inner part was subjected to accelerated carbonation resistance testing, allowing the assessment of natural and accelerated carbonation resistance on the same specimen.

The carbonation resistance was characterised by means of a coefficient given by the ratio between carbonation depth and the square root of exposure time. According to the literature on the subject a linear relationship between accelerated and natural carbonation coefficients, not influenced by cement type, was assumed. A complementary experimental study was carried out to investigate how age influences accelerated carbonation resistance.

Different factors, according to exposure classes, were determined by linear regression analysis to correlate accelerated and natural carbonation coefficients. The use of these factors together with Fick's first law enables the long term prediction of carbonation depth in structures based on accelerated carbonation resistance testing.

1. Introduction

1.1. Preliminary remarks

Concrete carbonation is a physicochemical process that involves the diffusion of CO_2 through the concrete pores and its reaction with hydration products such as calcium hydroxide and calcium silicate hydrates, a process that changes the concrete's physical and chemical properties. Although these reactions may have a beneficial impact [1–3], the main effect of carbonation in reinforced concrete structures is to lower the pH in the interstitial solution.

Non-carbonated concrete protects the steel reinforcement from corrosion as it provides a very alkaline environment ($pH \approx 13$), where the thin protective film of iron oxides around steel reinforcements is stable. Once pH falls to about 9 [4], steel reinforcement is no longer passivated and it becomes susceptible to corrosion.

Because the number of reinforced concrete structures suffering corrosion problems has increased significantly in recent years, leading to costly repair and strengthening works [5], carbonation, which, along with chloride penetration, is one of its major causes, has been the subject of interest, particularly in terms of the progression of carbonation over time.

1.2. Problem statement

The time it takes the carbonation front to reach the steel reinforcement is known as initiation period [6], and it is an important stage of the service life of reinforced concrete structures. Several methods to predict this period have been proposed, including analytical models (e.g. [7]), numerical models (e.g. [8]) and even neural networks [9].

Modelling carbonation is a delicate issue since although several factors should be taken into account users expect research to deliver simple and user-friendly models. Factors affecting carbonation can be organised into two major groups: environmental factors and factors intrinsic to concrete. The environmental factors include: occurrence of dry–wet cycles [10], relative humidity [11] and CO_2 concentration [11]. Intrinsic factors are mainly related to concrete's porous structure, which influences its mass transport properties, and to the amount of the substance that can be transformed into carbonates.

The process of CO_2 diffusion can be modelled based on Fick's first law. The initial model to predict carbonation depth, proposed by Tuutti [6], is based on the diffusion law and considers that the

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carbonation rate is proportional to the square root of the time of exposure to the CO_2:

$$x = K\sqrt{t} \tag{1}$$

where x is the carbonation depth (mm); K is carbonation coefficient (mm/year$^{1/2}$); and t is exposure time to CO_2 (year).

The carbonation coefficient summarises several parameters, assumed to be constant: CO_2 concentration, amount of CO_2 required to carbonate a unit volume of concrete, and diffusion coefficient for CO_2 through carbonated concrete.

However, in real exposure conditions, CO_2 concentration is not constant over time [12], the diffusion coefficient is sensitive to relative humidity and dry–wet cycles and the amount of CO_2 required to carbonate a unit volume of concrete depends on the amount of CO_2 available inside it [13], because for regular CO_2 concentrations only the hydrated cement compounds react with CO_2, while for high CO_2 concentrations these and the anhydrous cement compounds both react with it [14]. Some models [15–20] consider the influence of dry–wet cycles, modifying the exponent of time to less than 0.5 as its action slows the carbonation rate. Other models [7,21–25] incorporate the effect of dry–wet cycles into the carbonation coefficient. Actually, when the carbonation depth is taken to be proportional to the square root of time the carbonation coefficient will reflect the influence of all the other factors.

Under the same environmental conditions, including CO_2 concentration, the coefficient K is a relative indicator of concrete carbonation resistance and the most straightforward and reliable parameter for characterising the influence of concrete intrinsic factors on carbonation. To evaluate carbonation resistance and collect results in good time, accelerated tests have to be used in which concrete is exposed to atmospheres of relatively high CO_2 concentration.

Even though results from accelerated carbonation tests have been used to predict long-term carbonation depths [18,19], there is no general agreement on how to extrapolate from accelerated testing conditions to real performance [26–28]. This is indeed a complex matter as the mass transport properties of laboratory specimens and structural elements differ, and the CO_2 concentration of accelerated testing and natural exposure conditions also differ.

Mass transport property disparities arise from different placing, compacting and curing, and from relative humidity and dry–wet cycles. These last two factors may also affect the chemical part of the carbonation process.

Although, according to Fick's first law, the carbonation depth is proportional to the square root of CO_2 concentration, some studies [13,29,30] have shown that there are other factors in the kinetics of carbonation chemical reactions whose influence may invalidate the above-referred relationship [31].

1.3. Objectives and scope of the research

The aim of this study is to investigate carbonation resistance under accelerated and under natural conditions as a contribution to a more reliable prediction of concrete carbonation progression in structures using laboratory accelerated test results.

Major drawbacks in existing studies on the subject are the use of laboratory specimens to assess carbonation resistance in natural conditions, scarce data preventing statistical inference and assessment of carbonation resistance in relatively short periods of exposure to natural conditions. These were overcome by assessing carbonation resistance in real structures in 96 locations; different structural elements, ranging in age from 4 to 32 years, were examined.

1.4. Method

The method involved the following steps:

- Assessment of carbonation resistance in natural and accelerated conditions using two sides of the same sample, extracted from an existing structure.
- Quantification of carbonation resistance under accelerated and natural conditions, through carbonation coefficients.
- Evaluation of the influence of the age at testing on accelerated carbonation resistance, using laboratory specimens, as tested field specimens were of aged concrete, whereas in current practice accelerated carbonation resistance must be assessed at around 28 days.
- Using statistical tests to investigate the influence of age and environmental conditions on the ratio between accelerated and natural carbonation coefficients.
- Selection of a general relationship between accelerated and natural carbonation coefficients, supported by a literature review.
- Investigation of influence of cement type on the selected relationship.
- Regression analysis performed to establish specific relationships between accelerated and natural carbonation coefficients, according to exposure classes.
- Comparison of the computed values with results from other studies.
- Discussion of the implications of using different CO_2 concentrations in accelerated carbonation testing based on existing studies on the subject.

2. Experimental programme

The experimental program assessed carbonation depth in cores drilled from the surface of real structures that were exposed to natural conditions. The non-carbonated inner part of the cores was subjected to accelerated carbonation resistance testing (Fig. 1). Cores were taken from structures which were in service and from structures about to be demolished. For technical reasons, e.g. power supply, cores were taken from in-service structures using a core bit with a diameter of 50 mm, the minimum recommended in EN 14630 [32] whenever maximum aggregate size is over 16 mm. In the structures about to be demolished, 75 mm diameter cores were taken from larger cores provided by the holes drilled during the demolition process. Additional tests were also run under accelerated conditions on laboratory specimens, cylindrical slices, 50 mm thick, sawn from 150 mm diameter and 300 mm tall cylinders.

2.1. Measurement of carbonation depth

The carbonation front depth was determined by means of a spray with a solution of 1% phenolphthalein in 70% ethyl alcohol,

Fig. 1. Cross-section of a split core with carbonation fronts from natural carbonation (left-hand side) and accelerated carbonation (right-hand side).

which changes colour (from dark rose to colourless[3]) and thus indicates the concrete areas with a pH below 9, which was the region where the natural alkalinity of the concrete had been neutralised (Fig. 1).

The depth of carbonation in a freshly broken surface was measured by means of a calliper rule with a device at its end to facilitate the visualisation of the reference position of the concrete surface, using the precision suggested in RILEM recommendation CPC-18 [33].

An average of 9 readings of carbonation depth were performed for each carbonation front and the respective arithmetic mean was taken as its representative value, while for laboratory specimens the representative value was the arithmetic mean of 24 readings.

2.2. Natural conditions

The carbonation resistance under natural conditions was assessed in the uncoated original surfaces of 21 viaducts, between 4 and 32 years old, located within a radius of 50 km from Lisbon and at least 2 km from the coast. Columns, decks and abutments were assessed. The samples from columns and abutments were taken from vertical surfaces, while the samples from decks were taken mainly from bottom faces of slabs but also from lateral faces of precast beams.

Regarding the exposure conditions, EN 206-1 [34] defines 4 exposure classes for carbonation induced corrosion: XC1, XC2, XC3 and XC4. The XC1 exposure class stands for permanently dry (e.g. buildings' interior) or permanently wet (e.g. totally immersed) concrete. The XC2 exposure class stands for concrete with long periods in contact with water (e.g. rainwater drainage systems). Environments with moderate humidity (e.g. concrete in open air structures sheltered from rain) correspond to exposure class XC3 and with dry–wet cycles (e.g. concrete in open air structures not sheltered from rain) correspond to exposure class XC4. According to these definitions two cores were taken from surfaces exposed to class XC2 environmental conditions, 65 cores were taken from surfaces exposed to class XC3 environmental conditions and 29 cores were taken from surfaces exposed to class XC4 environmental conditions. As only a very small number of results were determined for XC2 class environmental conditions, they were not considered for further analysis. Some viaducts were in urban areas and others were in less polluted areas.

2.3. Accelerated conditions

The accelerated carbonation testing was performed on the non-carbonated inner part of the cores extracted.

A regular surface orthogonal to the core axis was achieved by sawing the inner edge. Then cores were kept inside a climatic chamber at 20 °C and 65% RH for 21 days, following the Lay and Schieβl [35] procedure. Afterwards, the lateral surfaces of the cores and the flat already naturally carbonated surface were coated with epoxy paint (50 mm diameter cores) or with self-adhesive aluminium tape (75 mm diameter cores) and wrapped in polyethylene film before being returned to the climatic chamber for 7 more days.

The cores were then placed inside a carbonation chamber at 20 °C and 65% relative humidity (RH), as suggested in [33], with a controlled 5% CO_2 concentration, as specified in the Portuguese standard for accelerated carbonation testing [36].

The regular exposure period under accelerated conditions was 28 days, although shorter exposure periods were considered in cores with small non-carbonated depth, aiming to avoid the overlapping of natural and accelerated carbonation fronts.

As resistance to accelerated carbonation was assessed in aged concrete, a parallel study to investigate the influence of concrete age on carbonation resistance was developed.

Specimens of 11 mixes using three types of cement: CEM I, CEM II/A and CEM IV/A, with water/cement ratios ranging from 0.39 to 0.61, were tested for accelerated carbonation resistance at 5 weeks and at older ages (between 1 and 4 years).

The original specimens, cylinders with a diameter of 150 mm and height of 300 mm, were removed from moulds at 24 h and kept in tepid (20 °C) water for 7 days. They were then sawn into 50 mm thick slices and their moulded (curved) surface was coated with a double layer of adhesive aluminium foil. After that they were kept in the same climatic chamber as the cores until testing. The specimens due to be tested at older ages were wrapped in polyethylene film after spending 4 weeks inside the climatic chamber.

The accelerated carbonation tests were conducted in the same carbonation chamber containing the cores. A specimen from each mix was taken from the chamber at regular intervals and broken to measure the carbonation depth.

Detailed information about the concrete mixes and reasons for choosing the procedures adopted can be found elsewhere [37].

3. Results

The parameter selected to characterise carbonation resistance, under both natural and accelerated conditions, was the carbonation coefficient. This relates carbonation depth to the exposure period according to Eq. (1), as the latter has proven to be an adequate model for carbonation under natural [37–42] and also under accelerated [31,37,43–46] conditions.

3.1. Analysis

A preliminary analysis of the results showed that although the walls of abutments were exposed to XC4 environmental conditions their corresponding natural carbonation coefficients proved to be statistically different from the other natural carbonation coefficients (K_n) obtained for that exposure class. When preparing specimens for accelerated carbonation testing, in some of them thin cracks visible with naked eye were detected when their surface was drying. These specimens ended up exhibiting higher accelerated carbonation coefficients (K_a) than the average of the rest. Finally, the preliminary analysis also raises doubts about the real concrete age in two of the viaducts.

Therefore, the results from all the identified situations, together with the results from concrete exposed to class XC2 environmental conditions (see Section 2.2), were disregarded in the subsequent analysis, which left 60 pairs to support the present investigation. They are presented in Fig. 2.

First, the influence of the natural exposure time on the relationship between carbonation resistance under natural and accelerated conditions was evaluated. For this an analysis of variance (ANOVA) [47] was performed, considering three groups of ages: 10, 20 and 30 years, with 7, 14 and 39 elements, respectively.

On completion of the test, the null hypothesis was that the expected values of the accelerated carbonation coefficient-natural carbonation coefficient ratios (K_a/K_n) would be identical for all groups and, as an alternative hypothesis, the expected values of K_a/K_n were not all identical. The *p*-value, i.e. the significance level which allows the acceptance of the null hypothesis, was 0.42. Assuming a significance level of 5%, the null hypothesis prevailed,

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[3] For interpretation of colour in Fig. 1, the reader is referred to the web version of this article.

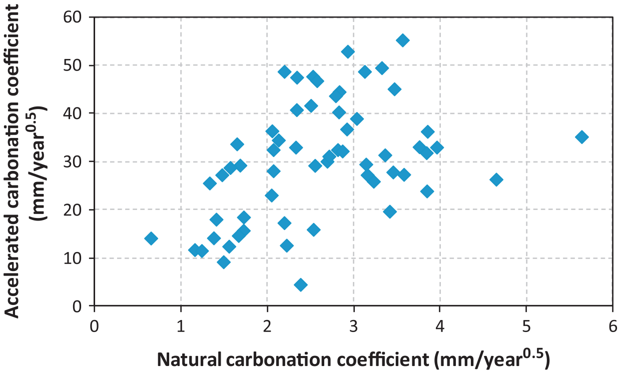

Fig. 2. Natural carbonation coefficient vs. accelerated carbonation coefficient (overall results).

Table 1
Statistics of K_a/K_n.

Group	Number of elements	Mean of K_a/K_n	Standard deviation of K_a/K_n
XC3-rural	36	11.1	4.7
XC3-urban	9	11.7	4.3
XC4-rural	3	13.3	5.2
XC4-urban	12	16.5	3.0

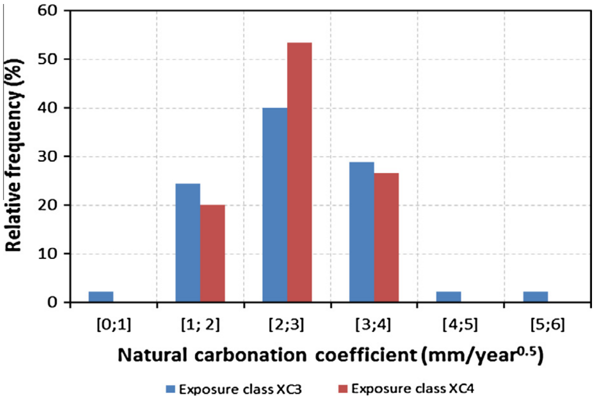

Fig. 3. Relative frequency histograms of K_n.

indicating that there is no age influence on the ratios obtained in the different groups.

Given the influence of relative humidity and dry–wet cycles on the carbonation rate [48,49] and the different carbonation rates for the same concrete, exposed to natural environments with and without additional sources of CO_2, reported by Saetta and Vitaliani [50], the results were grouped by exposure class and any presence of additional sources of CO_2. Four groups were considered: XC3-urban, XC3-rural, XC4-urban and XC4-rural (Table 1).

Although it would be worth comparing XC4-urban with XC4-rural and XC3-rural with XC4-rural, that was not possible as the XC4-rural group was held to be not representative due to the very small number of results. Therefore only the XC3-urban and XC3-rural, and XC3-urban and XC4-urban groups were compared. The mean value of K_a/K_n for XC3-urban was higher than for XC3-rural, contrary to what might be expected, which can only be explained by statistical uncertainty. The statistical significance of the difference between mean values of K_a/K_n for the XC3-urban and XC4-urban groups was assessed using Student's *t*-test [51]. On completion

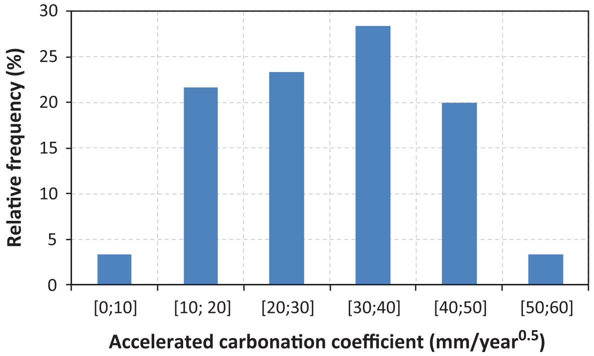

Fig. 4. Relative frequency histogram of K_a.

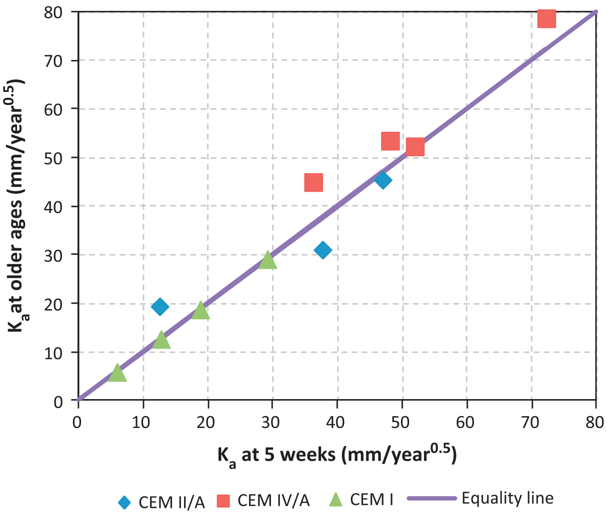

Fig. 5. Comparison of accelerated carbonation resistance at 5 weeks and at older ages.

of the test, the null hypothesis was that the expected values of K_a/K_n were identical in both groups and, as an alternative hypothesis, the XC4-urban group's expected value of K_a/K_n would be higher than the XC3-urban group's expected value of K_a/K_n. The *p*-value was 0.003. Assuming a significance level of 5%, the null hypothesis was rejected, demonstrating the influence of the environmental conditions on the carbonation rate.

As no clear evidence of any additional sources of CO_2 was found in the present investigation, and nor was any found by Neves [37] in a similar investigation that considered only natural carbonation coefficients, the results are only grouped by exposure class in the discussion below. It is noted that the difference in carbonation rates due to additional sources of CO_2 reported by Saetta and Vitaliani [50] occurred in closed environments with the additional source on the inside.

The mean value and standard deviation for K_n for the XC3 class were respectively 2.63 mm/year$^{0.5}$ and 1.01 mm/year$^{0.5}$, while for the XC4 class they were 2.45 mm/year$^{0.5}$ and 0.69 mm/year$^{0.5}$, respectively. Relative frequency histograms of K_n for the two environmental exposure classes are presented in Fig. 3. A relative frequency histogram for the corresponding laboratory accelerated testing results is presented in Fig. 4. The mean value and standard deviation of K_a were 30.3 mm/year$^{0.5}$ and 12.1 mm/year$^{0.5}$, respectively.

The results of accelerated carbonation resistance testing in laboratory specimens are presented in Fig. 5. Despite the apparent absence of age influence on carbonation resistance from 5 weeks forward, a Student's paired two-sample *t*-test [47] was performed. On completion of the test, the null hypothesis postulated a null difference between K_a determined in tests at 5 weeks and at older ages and, as an alternative hypothesis, a non-zero difference was postulated. The *p*-value was 0.29. Assuming a significance level of 5%, the null hypothesis is accepted, thus confirming that age does not influence carbonation resistance from 5 weeks forward.

4. Discussion

From a literature review [14,51,52] it seems plausible to assume a linear relationship between K_a and K_n. Moreover, a linear relationship is already considered in reference carbonation models [18,19].

The complementary tests showed no influence of age on accelerated carbonation resistance from 5 weeks forward.

As no information was found on the field concrete mixes it was not possible to ascertain the type of cement in the concrete of the tested cores. Given the Portuguese construction practices at the time when the viaducts were built, it was presumably cement CEM I. A literature search was carried out to investigate the influence of cement type on the relationship between K_a and K_n. Looking at the accelerated carbonation results presented in [23] along with the natural carbonation results presented in [53], which actually belong to the same research project, there is no evidence that cement type influences the relationship between K_a and K_n (Fig. 6). The same inference can be drawn from results presented by Gehlen [52] and Hamada's results [54] compiled by Parrott [14]. Fig. 6 also reinforces the thesis of a linear relationship between K_a and K_n and confirms the influence of environmental exposure on that relationship.

Assuming a linear relationship between K_a and K_n, the influence of the environmental exposure conditions and that the relationship is the same regardless of the type of cement, a regression analysis [47] was performed for the groups of results reported in the previous section. The low correlation coefficients obtained, 0.23 for XC3 group and 0.50 for XC4 group, led to a confidence interval being calculated for each of the relationships (Figs. 7 and 8). The estimated slope, also known as acceleration factor, the limits of its 95% confidence interval and the standard error of each regression are presented in Table 2.

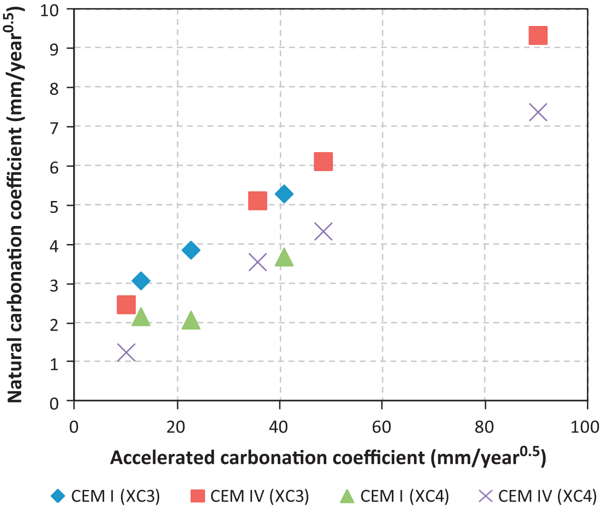

Fig. 6. Cement-type influence on the relationship between K_a and K_n (data from [23] and [53]).

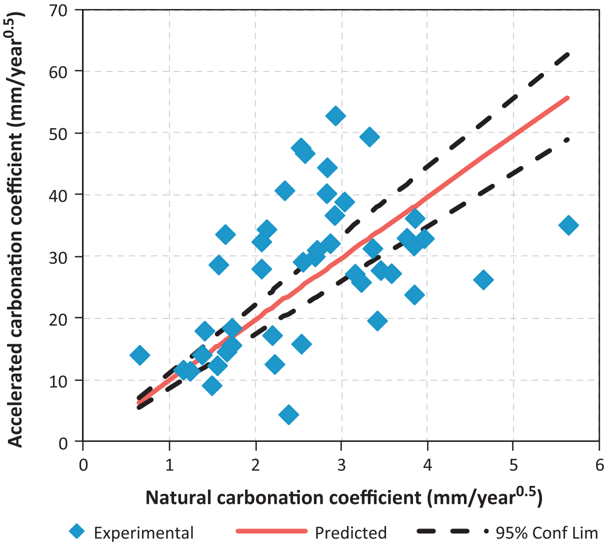

Fig. 7. Relationship between natural and accelerated carbonation coefficients for XC3 exposure class.

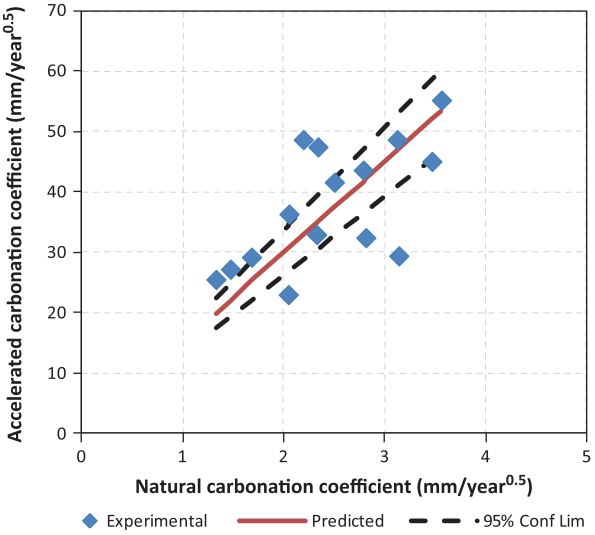

Fig. 8. Relationship between natural and accelerated carbonation coefficients for XC4 exposure class.

Table 2
Regression analysis results.

Acceleration factor	Exposure class XC3	Exposure class XC4
Estimated	9.9	15.0
95% Confidence interval	[8.7;11.1]	[13.1;16.9]
Standard error	0.6	0.9

The low correlations are due to the high scatter of the results, which is attributed to the diversity of situations, e.g. stress level [55], within the same exposure class. There is actually a low scatter in Fig. 6, as both carbonation resistances were assessed in laboratory-moulded specimens and natural exposure conditions were exactly the same for all specimens.

Most of the studies on the relationship between carbonation resistance under natural and under accelerated conditions chose an indoor (laboratory) environment, equivalent to a XC1 exposure class, for natural exposure conditions. These investigations led to acceleration factors approaching 7.5 [27,51,56–58], even though

CO_2 concentration in accelerated testing varied between 4% and 10%. Pauletti [58] exposed mortar specimens to accelerated (CO_2 concentration of 6%) and natural (XC4 and subtropical climate) conditions and obtained a mean acceleration factor of 11.3. Khunthongkeaw et al. [59] exposed concrete and mortar specimens to an accelerated (CO_2 concentration of 4%) and a natural tropical climate, XC3 and XC4 conditions, and obtained acceleration factors around 9 and 10 for XC3 and XC4, respectively. Applying a regression analysis to the results from [23] and [53], presented in Fig. 6, where CO_2 concentration in accelerated tests was 5%, and where natural exposure was to a Mediterranean climate, acceleration factors of 11.9 and 13.4 for XC3 and XC4 exposure classes, respectively, are obtained.

Therefore, the application of Eq. (1) together with the information presented in Table 2 is believed to be a suitable method to predict long-term concrete carbonation in structures. However, potential users must be aware that the proposed acceleration factors were obtained under specific conditions, with special emphasis on the CO_2 concentration of the accelerated testing. Many authors [7,20,24,55], including reference models [18,19], assume carbonation depth to be proportional to the square root of CO_2 concentration, while others do not [27,31]. According to the experimental results of Hyvert et al. [31], where a large range of CO_2 concentrations were considered, the relationship between carbonation depth and CO_2 concentration deviates markedly from a linear relationship. Actually, even in a rough approach at least a bilinear relationship should be considered. Nevertheless, when using CO_2 concentrations of near 5% in accelerated testing for carbonation resistance, a correction of the acceleration factor using the square root of accelerated testing concentrations ratio is a reasonable option so as to still be able to use Eq. (1) and Table 2 information to predict long term concrete carbonation progression in structures. When using higher CO_2 concentrations, e.g. 20%, other acceleration factors should be contemplated.

5. Conclusions

Cores were taken from real structures and their non-carbonated inner part was subjected to accelerated carbonation resistance testing, allowing the assessment of natural and accelerated carbonation resistance, characterised by means of a coefficient of carbonation, given by the ratio between carbonation depth and the square root of exposure time, on the same specimen. Field results allowed the investigation, with statistical inference, of the relationship between natural and accelerated carbonation resistance for exposure classes XC3 and XC4.

To support the main investigation of the present study, some subjects had to be discussed. The evaluation of the influence of age and environmental conditions on the ratio between accelerated and natural carbonation coefficients was performed using the results obtained from core samples. It was concluded that age (length of exposure period to natural conditions) has no influence, while the environmental conditions influence that ratio. This finding prompted the consideration of two groups, in terms of exposure class (XC3 and XC4). The evaluation of the influence of age on accelerated carbonation resistance testing was performed using laboratory specimens especially prepared for this purpose and led to conclude that age does not influence carbonation resistance from 5 weeks forward. Based on a literature review a linear relationship between natural and accelerated carbonation coefficients was adopted and it was also found that the type of cement has no influence on that relationship.

Despite the high scatter of the results, a simple method that may include probabilistic concepts is proposed to predict long term concrete carbonation in structures. The method involves the use of Fick's first law (Eq. (1)) and accelerated carbonation resistance test results (K_a). The latter must be corrected deterministically or probabilistically by an acceleration factor (a) which considers the influence of curing and service conditions, especially the environmental exposure conditions, i.e. $K = K_a/a$. The acceleration factors determined by this investigation were compared to values found in other studies and differences were justified.

When CO_2 concentrations that differ slightly from 5% are used in accelerated carbonation resistance testing, a correction of the acceleration factor by the square root of accelerated testing concentrations ratio is suggested.

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