Towards Practical Carbonation Prediction and Modelling for Service Life Design of Reinforced Concrete Structures

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Abstract. Amongst the scientific community, the interest in durability of concrete structures has been high for quite a long time of over 40 years. Of the various causes of degradation of concrete structures, corrosion is the most widespread durability problem and carbonation is one of the two causes of steel reinforcement corrosion. While much scientific understanding has been gained from the numerous carbonation studies undertaken over the past years, it is still presently not possible to accurately predict carbonation and apply it in design of structures. This underscores the complex nature of the mechanisms as influenced by several interactive factors. Based on critical literature and some experience of the author, it is found that there still exist major challenges in establishing a mathematical constitutive relation for realistic carbonation prediction. While most current models employ permeability/diffusion as the main model property, analysis shows that the most practical material property would be compressive strength, which has a low coefficient of variation of 20% compared to 30 to 50% for permeability. This important characteristic of compressive strength, combined with its merit of simplicity and data availability at all stages of a structure’s life, promote its potential use in modelling over permeability. By using compressive strength in carbonation prediction, the need for accelerated testing and permeability measurement can be avoided. This paper attempts to examine the issues associated with carbonation prediction, which could underlie the current lack of a sound established prediction method. Suggestions are then made for possible employment of different or alternative approaches.

1. Introduction
Carbonation as a mechanism of deterioration has increasingly become an important worldwide problem in concrete structures. This is due to the fact that corrosion of steel reinforcement is the most widespread cause of deterioration in concrete structures. This raises the need for two sets of actions, consisting of: (a) durability specification, (b) service life design. Specification for durability requires understanding of the attack mechanisms and employment of measures that mitigate the attack process. Obviously, this requires knowledge of the attack process and the investigation of the material systems that are capable of mitigation the attack. This segment of understanding carbonation has been well-
established by researchers over the past decades. For example, it is known that extenders generally tend to increase the progression of carbonation in concrete and that higher concrete strength reduces carbonation progression. With the present understanding of the carbonation process, durability specification for carbonation resistance is possible and is being practiced. But it is the development of a service life method of design for carbonation that has eluded researchers for quite a long time, despite the concerted efforts by the scientific community. A literature review by Parrot [1] and Sagues et al. [2] show that not only there are several factors that influence carbonation but synchronising the effects of all these factors into a prediction model comes with extremely intriguing tasks that continue to be investigated by the scientific community.

To date, a myriad of carbonation models have been proposed in the literature, which indicates the high level of interest and enthusiasm in carbonation studies. But it is often the case that prediction models developed on the basis of a particular data set tend to fail when applied to another independent data source. This paper attempts to identify the reasons for lack of practical carbonation prediction models and articulates on the important considerations in developing prediction models.

2. A future of carbonation

2.1. Historical perspective
Carbonation of concrete structures is a direct result of the increase in green house gas emissions in the Earth’s atmosphere, which has emanated from the era of Industrial Revolution believed to have occurred from about 1750 to 1850. This event, which in terms of changes to human living, is often considered to be one of the most important eras in human history, was a period when production process shifted from the use of basic machines and hand tools to power-driven machinery which allows mass production. The emergence of technological advances over the hundred year period of Industrial Revolution did change lives dramatically as factories opened and workers congregated to locations of industrial production, which eventually turned into urban centres or cities. After 1860’s a new drive was infused in what is often referred to as the Second Industrial Revolution, when scientists and engineers began to employ scientific approaches based on physics and chemistry in order to unlock industrial practices, which in turn led to further discoveries and has still continued to the present age.

To a great extent, the Industrial Revolution is hailed as the period of great transformation to improve living conditions, but along with its benefits it brought about huge problems which continue to this day. These include environmental hazards from toxic waste, atmospheric pollution from factory gas emissions, health problems for factory workers, diseases from waste disposal, poor sanitation, etc., which continue to be major modern challenges to the industry. The chain of success in industrialisation, however, is energy driven. Before the Industrial Revolution, human physical effort was the main energy source for machine tools and other forms of industrial work. This was replaced by steam engine power at the initial stages of the revolution, followed by the emergence of petroleum oil and electrical power in the 19th century and currently these remain the mainstream sources of industrial energy in the modern industry.

2.2. Future trends
Figure 1 shows that the growth in CO₂ emissions commenced from onset of the industrial revolution and intensified as the second wave of the revolution spread worldwide creating massive and more intense industrialisation as a means to economic sustainability of the growing populace. Another
implication of CO\textsubscript{2} emission is climate change effect, believed to be the leading cause of global warming. In the reinforced concrete structures, CO\textsubscript{2} is responsible for steel corrosion which causes major repair and maintenance issues. As seen in figure 1, this trend of human co-existence with CO\textsubscript{2} emissions is set to continue as long as industrialization exists in its current form of using fossil fuels. Even more crucial is the projected trend showing possible rapid or exponential increase in CO\textsubscript{2} emissions in the near future. While international efforts are presently being summoned to reduce CO\textsubscript{2} emission, it remains a major problem that the industry will contend with far into the future, provided fossil fuels remain the primary source of energy for industrial processes.

![Figure 1. Global trends in CO\textsubscript{2} emissions; (a) Traceability to Industrial Revolution; (b) Emission by fuel type [4-6].](image)

3. Theoretical expression of carbonation
The fundamental law governing CO\textsubscript{2} diffusion into concrete is well-established to be the Fick’s first law, written as

\[ J = D \frac{dC}{dx} \]

where \( J \) is the amount of ionic mass agent (CO\textsubscript{2} in this case) passing through a unit area (A) of a porous medium over a unit period of time (t), \( dC/dx \) is the concentration gradient between the surface and the interior, and constitutes the driving force of CO\textsubscript{2} ingress, D is the diffusion coefficient of the material i.e. concrete in this case, which is a property dependent on connectivity of the pore network in concrete. Application of this theoretical physical law to carbonation in concrete leads to the expression of CO\textsubscript{2} ingress into concrete as a square root function of time, given as [7,8]:

\[ x = K \sqrt{t} \], where K is the carbonation rate coefficient given as \( K = \left( \frac{2D(C_1 - C_2)}{a} \right)^{\frac{1}{2}} \)

\( D_c \) is CO\textsubscript{2} effective diffusion coefficient, \( C_1 - C_2 \) is concentration difference between CO\textsubscript{2} in air and carbonation front, \( a \) – alkaline content in the concrete. Models attempt to accurately predict the carbonation rate coefficient and to do so consistently for various types of concretes subject to different environmental exposure conditions. It is at this point where the complexity of developing or building prediction models unfolds.
4. Carbonation prediction and modelling approaches
Models attempt to predict carbonation in concrete by mathematically or empirically relating the progression of carbonation to a major property of the concrete material system. Theoretically, diffusion or permeability would naturally be the material property of interest in this regard. However, many models proposed on this basis have failed to provide consistent, sound and realistic carbonation prediction. Accordingly, researchers have come to realise that there are many other properties of concrete that relate strongly to carbonation. Through experimental studies, numerous forms of models have been proposed including empirical and mathematical models [1,2], kinetic or physicochemical models such as [10] that are based on chemical reaction determinants, statistical models such as [11], which typically employ multiple regression of various parameters to obtain predictive relationships, simulations and artificial neural networks such as [12], numerical models such as [13,14], which employ computational techniques that are heavily computer reliant. Of these model types, it is the empirical-mathematical models that often find preferred acceptance in design of engineering structures. Models of this kind have been developed in the past 50 years for creep and shrinkage phenomena and used in established international design codes such as ACI 209, BS 8110, RILEM B3, Wendner et al., 2013, CEB-FIP 1978, 1990; AS3600, SANS 10100 [15-22]. The empirical-mathematical models are admired for their universal veracity and realistic predictions with relatively less complexity as they tend to be of practical inclination. In carbonation studies, no such models have been fully developed and very few models, such as Duracrete [23], have been proposed as experimental models for practical carbonation prediction. In this paper, an attempt is made to highlight the potential sources of problems in the development of empirical-mathematical models for practical carbonation prediction.

In this work, selection of the empirical-mathematical models in the category of potential practical viability is made. However, in consideration of ease for practical application, it is essential to examine the parameters that are involved in the model function, i.e. whether they contain the merit of simplicity, whether the data for their use is easily available or can be easily acquired in a typical design project setting. These criteria lead to elimination of some models, such as those that require the use of mix composition, i.e. water-cement ratio (w/c) or cement content [1], since it is hardly possible to accurately determine these parameters in the hardened concrete. The CEB-FIP model [24], for example, is non-committal on recommending any specific performance property but suggests examples of using mix parameters (such as w/c, cement type etc) while recommending the use of experimental data and field testing to derive the carbonation rates. But the model also gives special consideration to the influence of exposure environment by incorporating the weather function. Following an extensive literature review, models that appeared to be oriented to these two criteria of empirical-mathematical functionality and potential practical applicability were selected as shown in table 1. These models currently represent arguably the foremost empirical-mathematical carbonation models proposed in the literature.

5. Discussion of problems in carbonation modelling
5.1. Selection of a suitable property of concrete for constitutive relations
It can be seen in table 1 that the two main properties of concrete that feature highly in mathematical functions to relate carbonation progression are permeability/diffusion or concrete strength. In fact, the majority of the models [8,10,23-26] use permeability/diffusion as their constitutive property. However, problems arise in considering its implementation, which renders its use highly questionable.
Table 1. Mathematical prediction models for carbonation, selected based on functional and material performance parameters.

| Performance property | Prediction Model (d or A = carbonation depth) | Source |
|-----------------------|-----------------------------------------------|--------|
| Permeability          | \( d = \frac{ak^{0.4}t_n}{c^{0.5}} \), \( k \) = permeability, \( c = \text{CaO content}, a = 64 \) | Parrot [25] |
| CO₂ diffusion         | \( d = \frac{2C_o}{C_o + 1-e} \sqrt{\frac{D_{\text{eff}}}{\pi e}} \) \( C_o \) - Initial CO₂ concentration, \( D_{\text{eff}} \) - effective diffusivity coefficient, \( e \) - porosity | Xu and Rodhe [26] |
|                       | \( d = \sqrt{\frac{2D_c(C_1-C_2)}{a}} \) \( D_c \) - CO₂ effective diffusion coefficient, \( C_1-C_2 \) - concentration difference CO₂ in air and carbonation front, \( a \) - alkaline content in the concrete | CEB [8] |
|                       | \( d = \left( \frac{2[CO_2]}{D_{\text{eff}}} \right)^{1/2} \) \( \text{Deff} \) - effective diffusivity of CO₂, [CH], [CSH] are molar concentrations | Papadakis et al. [10] |
|                       | \( A = \sqrt{2e_{\text{env}}.D_{ca}} \) | DURACRETE /fib [23,24] |
|                       | \( D_{ca} = k_{\text{c,ca}} k_{\text{e,ca}} D_{e,ca} \left( \frac{t_o}{t+1} \right)^{2n_{ca}} \) \( D_{ca} \) - carbonation rate from the laboratory test, \( k_{\text{c,ca}} \) - environmental factor, \( k_{\text{e,ca}} \) - curing factor, \( t_o \) - age of concrete during laboratory carbonation test, \( n_{ca} \) - age factor | |
| Concrete strength     | \( d = \frac{150ckd_{\text{f}}}{f_c} \) \( c \) - the parameter for CO₂ binding capacity, \( d \) - CO₂ surface concentration, \( f_c \) - compressive strength of concrete, \( k \) - the RH parameter | Bob [27,28] |
|                       | \( d = C_{\text{env}}C_{\text{air}}af_{ca}(f_{ca} + 8)^a \sqrt{t} \) \( t \) - time of exposure (years), \( C_{\text{env}} \) - environmental coefficient, \( C_{\text{air}} \) - air content coefficient, \( f_{ca} \) - characteristic cube compressive strength, \( a,b \) - constants dependant on binder type | Hakkinen [29,30] |
|                       | \( d = (A - B\sqrt{f_{ca}}).\sqrt{t} \) \( A,B \) constants allows for different types of concrete materials and age effect on strength | Kokubu, and Nagatakis [31] |
|                       | \( d = 680(f_{ca} + 25)^{2.5} - 0.6 \) | de Fontenay [1,32] |
Research has shown that permeability/diffusion property measurement is not sufficiently reproducible to be considered reliable. This also is the reason why there are no plausible standardized test methods for permeability that are universally recognised. The permeability property characteristically displays high variability, regardless of the kind of test method applied. This observation was highlighted by Hooton [33], who reported that the coefficient of variation (CV) for permeability varied from about 30% for oxygen permeability up to 50% for water permeability. The findings of Stanish et al. [34] reported CV values of 23.7% to 53.9% for oxygen permeability, which are in agreement with [33] as shown in table 2.

**Table 2. Coefficient of variation for permeability or diffusion measurements.**

| Reference       | Description                                   | Coefficient of variation (%) |
|-----------------|-----------------------------------------------|------------------------------|
| Hooton [33]     | Water permeability; 36 tests x 3 replicates   | 51.9                         |
| Day et al. [33] | Oxygen permeability; 32 tests x 5 to 9 replicates | 30.6                         |
| Hope and Malholtra [33] | Water permeability; 5 tests x 4 replicates | 44.4                         |
| Stanish et al. [34] | Oxygen permeability                        | 23.7 to 53.9                |
| Imamoto et al. [35] | Air permeability; 16 tests                  | 33.1 to 34.2                |

**Table 3. Coefficient of variation for compressive strength measurements.**

| Reference       | Description                                   | Coefficient of variation (%) |
|-----------------|-----------------------------------------------|------------------------------|
| Shimizu [36]    | Built before 1961; 453 core tests, 52 buildings | 26.8                         |
|                 | Built between 1961-65; 1769 core tests, 173 buildings | 27.1                         |
|                 | Built between 1966-70; 2530 core tests, 260 buildings | 19.8                         |
|                 | Built between 1971-75; 3398 core tests, 344 buildings | 18.9                         |
|                 | Built between 1976-80; 2086 core tests, 244 buildings | 15.6                         |
|                 | Built after 1980; 178 core tests, 21 buildings | 15.6                         |
| Ekolu [37]      | 15 highway bridges and highways, 20 to 70 years of age | 17.0                         |
| Other literature [29] | Recommended by various sources              | 20.0                         |
Another concern of using permeability measurement property is the need for specialized test equipment. Permeability testing requires quite expensive equipment which includes the permeameter, conditioning chamber or oven, and pressure chamber. The expensive equipment also requires specialized training of technical staff. Altogether, these factors result in quite a major expense whether the testing is conducted in-house or through commercial laboratories. These issues can inhibit the potential use of permeability techniques in most developing countries, considering the lack of permeability equipment and skills for conducting the tests in these countries. In addition, the costs of installing such a facility may be unaffordable for typical or regular testing.

Conversely, compressive strength has a fairly low coefficient of variation, of about 20% as shown in table 3 showing reports from various literature sources.

Not only is compressive strength strongly related to carbonation as shown in figure 1, its data are also the most abundant set of test results typically acquired before and during construction. It is also simple to extract cores from existing structures for strength measurement. It can therefore be seen that the use of compressive strength in constitutional relations would provide significant advantages over the use of permeability. This is more important in the developing countries where the most basic tests that may be conducted in construction would be compressive strength tests.

\[ \text{Figure 2. Relationship between compressive strength and carbonation (constructed from [38]).} \]

5.2. Role of climatic regions and global geographical settings

It is often the case that the influence of climatic regions is not given much consideration in development of models. However, this factor plays a major role in considering practical model predictions. A crucial consideration is that most developing countries (Africa, Asia, South America) are located in the tropical regions, where carbonation tends pose a bigger problem than in the temperate climates (Europe, North America, Russia, etc.), where chloride attack is more dominant than carbonation. Accordingly, in the temperate climates, there may be sole carbonation ingress or combined carbonation/chloride attack in structures. Such models may need to give consideration to the synergy between the two mechanisms. In tropical countries, mainly sole carbonation progression
needs to be considered except along the coastal regions, where the risk of combined carbonation/chloride attack exists. The environmental factors are another key consideration. Whereas the relative humidity values in either region may vary from as low as 40%RH to as high as 80%RH, temperatures in the regions differ widely. While temperatures in the tropical regions are fairly steady at typically 18 to 30°C, those in the temperate regions may fall as low as -20°C in winter. Therefore, the models that may function properly in one climatic region may not directly apply to carbonation prediction in other climate types.

5.3. Accelerated tests versus natural exposure

Just about all diffusion-based models [8,10,23-26] depend on accelerated tests for determination of the diffusion coefficient to be applied in the model. However, there is always a problem that accelerated tests may not be able to accurately depict the variable field conditions and accordingly cannot accurately predict actual field behaviour of the structure under exposure to a natural phenomenon such as carbonation. Not only are the samples used in accelerated tests much smaller than the elements they represent in actual structures, they are also subjected to unnaturally severe test conditions such as high pressures during permeability or diffusion test, etc. At best, some kind of correlation may be established between accelerated tests and field results, but these correlations may change from one test set to another, making it difficult to obtain a universal relation. It is often the case that materials may fail accelerated tests but perform well under field conditions [39,40].

While there is a place for accelerated tests, specifically in studying behavioural characteristics from the theoretical perspective, it would be more appropriate to use carbonation data from natural carbonation exposure to assemble models for realistic carbonation prediction in structures. However, this option is immediately confronted with shortage or lack of data thereof, more so for long-term studies spanning several decades. This remains a challenge, although the approach of using the data from natural exposure is slowly gaining momentum [41].

6. Summary

The objective of this work was to identify some of the main problems affecting or hindering practical prediction of carbonation. While wide ranging approaches to carbonation prediction do exist, it seems that the development of a veracious constitutive relationship expressed mathematically remains a challenge. Many models allude to diffusion or permeability as the performance property to be used in a mathematical constitutive function, typically based on the theoretical Fick’s law. Realistically, however, the high variability of permeability/diffusion test procedures makes the use of permeability data in any manner to be quite questionable. Besides this, accelerated tests are the only form of measurement that can be used to obtain permeability data, which typically do not truly represent realistic behaviour of the structure under natural exposure. In the developing countries, the use of permeability is far from practical, due to the high expense and skill required to conduct these tests.

It appears that the most realistic methods for practical prediction of carbonation for service life design should be empirical-mathematical models that can easily be incorporated into design codes. In employing these models, their applicability should be made realistic by adopting a material property which not only exhibits carbonation progression effectively but does so while maintaining practical viability of the model. Compressive strength was found to be potentially an effective material property, given its low variability of about 20%, availability and ease of data acquisition at any age of the structure.
Designers may need to distinguish the applicability of carbonation prediction models based on climatic regions, with tropical regions requiring mainly sole carbonation models while temperate climates may need to consider the synergy between carbonation and other deterioration processes, which might lead to more complex models. This can be done while allowing for different environmental factors in the specific regions.

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