



Review Analytical prediction on carbonation depth between Empirical and Mathematical methods

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Abstract

Carbonation is a chemical reaction that occurs when carbon dioxide from the air reacts with calcium hydroxide in concrete, forming calcium carbonate. This reaction reduces the alkalinity of the concrete, which can lead to the corrosion of reinforcing steel and ultimately structural damage. Carbonation depth is a critical parameter that indicates the extent of carbonation in concrete. Analytical prediction of carbonation depth is important in determining the durability of concrete structures. This paper presents an abstract on analytical prediction of carbonation depth. The abstract discusses various analytical models that have been proposed for predicting carbonation depth in concrete, including empirical, theoretical, and probabilistic models. Empirical models are based on experimental data and have been widely used in practice. They are simple to use but have limited accuracy and applicability. Theoretical models, on the other hand, are based on the principles of chemistry and physics and provide a more accurate prediction of carbonation depth. However, they require detailed information on the material properties and environmental conditions, which may not always be available.

Keywords: Carbonation, Carbonation depth prediction, Durability.

1 Introduction

1.1 Background

Concrete is the most prominently used construction material that moulds into any desired shape. Though the commencement of the steel as a building material and the addition of mineral admixtures into concrete business has brought about many changes in construction techniques, the use of cement as a basic binder material has not dwindled, mainly because of its excellent water resisting property.

The fire remains one of the serious potential risks to most of the buildings and structures and for reducing this cause few methods are also done[22]. Addition of the admixtures to concrete the strength and durability of the concrete increases with various mix proportions are been done[23,24]. Replacement of the cement in the concrete also changes the strength parameters[25,26].

Concrete is a composite material that consists essentially of a binding medium within which are embedded particles or fragments of aggregate (ASTM C125) [1]. The binding material being cement, hardened concrete too is impervious to water provided the concrete has less porosity. The embedded particles that are commonly used are coarse and fine aggregates. The voids of coarse aggregates are filled by fine aggregates. Addition of the alccofine in the concrete gain strength was been observed in the concrete[27].

Cement is a finely grounded material that by itself does not possess any binding property, but on reaction with water (hydration) develops strength. Portland cement is most widely used hydraulic cement, produced by pulverizing clinkers consisting essentially of hydraulic calcium silicates, and a small amount of one or more forms of calcium sulphate as an inter-ground addition (ASTM C150)[2]. The construction business in India



consumes 400 million tons of concrete per year and will soon reach a billion-ton mark in less than 10 years (Gowda et al. (2011)). Sesimic behaviour of the concrete can be observed in the concrete and also if need addition of the several material can be done to increase its strength and durability[28].

Concrete despite its high strength and ability to resist the transport of water is susceptible to chemical attacks and deterioration. The ability of concrete to withstand the chemical attacks, weathering action, abrasion, or any sort of deterioration during its service life under a given set of conditions is called durability. The strength and durability of concrete depend on the properties of its ingredients, proportions of design mix, exposure conditions, and curing type and duration. The durability of concrete is influenced by both physical reasons and chemical reasons. The physical reasons for concrete deterioration can be broadly classified into surface wear and cracks. The surface wear is due to abrasion, erosion, and cavitation while the cracking is due to change in volume, structural loading, and exposure conditions (Mehta & Gerwick, 1982) [3]. And rapidly stiffness was observed check its durability and strength[31].

The chemical causes for deterioration of concrete generally, but not necessarily involve, chemical reactions between the aggressive agents in the environment and the ingredients of the cement paste. One of the reactions that influence the durability of concrete structures predominantly is corrosion of steel reinforcement in the concrete, which is mainly caused either because of chloride attack or the carbonation of concrete. Also, the porosity and interconnected permeable pores in concrete have a strong influence on its mechanical strength and durability (Kumar & Bhattacharjee (2003)) [4]. Experimental analysis was performed on the concrete to observe its strength and durability[29].

Carbonation of concrete is the reaction of calcium (Ca^{2+}) ions present in cement and carbonate (CO_3^{2-}) ions from the dissolved carbon dioxide (CO_2) to precipitate calcium carbonate (CaCO_3). Concrete possesses certain porosity when design mix proportion standards are not met. Also, upon prolonged exposure to the atmospheric fluids, the surface of concrete gets deteriorated and sometimes through its interconnected capillary pores, the deterioration progresses.

Carbonation of concrete is one such phenomenon where the surface of concrete gets exposed to the atmospheric carbon dioxide (0.03-0.04%) for a prolonged duration and changes the physicochemical properties of the cement hydration products. The carbon dioxide diffuses across the surface due to the difference in concentration between 3 atmosphere and concrete pore structure. A thin layer of carbonated concrete is formed initially, and the further diffusion of carbon dioxide depends on the permeability of the concrete (Mark, 2003). Addition of the geopolymers with the admixture it was observed the rapid strength and durability in the concrete[30].

Carbonation is a process where dissolved carbon dioxide from the atmosphere reacts with calcium ions in cement and form calcium carbonate. The solubility of calcium carbonate is less compared to calcium hydroxide, which eventually leads to the dissolution of all calcium compounds to calcium carbonate. The end products apart from calcite are silica gels and metal hydrates. Because carbon dioxide is abundantly found in the atmosphere and the cement used in construction purposes contains calcium hydroxides, the question is not whether the process of carbonation takes place, which is evident, but how long will this process take to initiate corrosion (Lagerblad, 2005) [5]. Prior to carbonation, the reinforcement in concrete is embedded in oxygenated alkaline solution. This deposits a thin layer of insoluble oxide film preventing oxygen from reacting with steel, which inhibits corrosion.

Due to the lowering of pH value during carbonation, the passive oxide layer gets destroyed and concrete acts as an electrolyte for oxygen and moisture to penetrate. This results in corrosion of reinforcement in reinforced concrete. This situation within the concrete has created a problem for the usage of steel as reinforcing medium. There is no dearth of cases where the corrosion of reinforcement has occurred even before the carbonation has reached the reinforcement as detected by phenolphthalein indicator (Yongsheng et al. 2010) [6].

This poses a question if the phenolphthalein indicator is an accurate measure of carbonation depth. Also, though well addressed, the effect of mineral admixtures with various pore sizes on carbonation resistance is seldom studied.

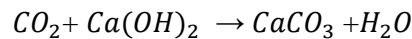


1.2 Chemistry of carbonation

The process of carbonation is a surface phenomenon and hence the amount of carbonation depends on the exposure duration and the porosity of the concrete. If the surface of the concrete is porous enough, the atmospheric carbon dioxide diffuses through the pores of surface concrete and dissolves in the pore solution of concrete to form carbonate ions due to the difference in concentration between the atmosphere and concrete pore structure.

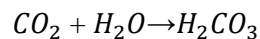
The gaseous carbon dioxide cannot react with the calcium phases of the cement. Based on the pH of the solution in which the carbon dioxide is dissolved, the carbonate ions are formed. When in contact with water at pH 7, bicarbonate ions are formed, but as the pH in the cement pore solution is high, the bicarbonates further disassociate to form carbonate ions.

Carbonation occurs when CO_2 , as natural gas in the air, penetrates the surface of concrete through the dry portions of interconnected pores in concrete microstructures, and reacts with $Ca(OH)_2$ in the moist portions of pores to form $CaCO_3$ and water (H_2O).

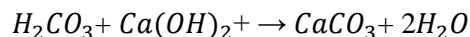


When $Ca(OH)_2$ from the paste is consumed, the hydration of calcium-silicate-hydrate (C-S-H) releases calcium oxide (CaO), which reacts with diffused CO_2 to form carbonates. The remaining CO_2 continues to react with C-S-H to form additional $CaCO_3$. The carbonation process requires water because CO_2 dissolves in water to form carbonic acid H_2CO_3 .

The water seeps in through small, interconnected pores, while CO_2 settles in large pores. The interconnected pores merge into large pores to supply the water required to form carbonic acid:



A chemical reaction occurs with H_2CO_3 , which is formed in the pores of the concrete, producing a desirable product of cement hydration $Ca(OH)_2$, which strengthens the concrete matrix. This reaction also produces $CaCO_3$ and water:



This process involves consumption of Ca^{2+} ions, which leads to the dissolution of predominantly available calcium hydroxide ($Ca(OH)_2$) forming new Ca^{2+} ions. Because the solubility coefficient of dissolution of $Ca(OH)_2$ is 9.95×10^{-4} mol/L and solubility coefficient of formation of $CaCO_3$ is 0.99×10^{-8} mol/L, this process continues till all the calcium ions are consumed to form $CaCO_3$ (Lager lad, 2005).

2. Overview of Empirical Carbonation Models Proposed

The model was proposed by Alekseev and Rozental (1976)[7] for the depth of carbonation prediction and also the relation between the depth of carbonation and time in accordance with water binder ratio.

$$x = A^n t$$

where



'A' is coefficient of carbonation

'n' = 1.92 for water/binder ratio 0.6

'n' = 2.54 for water/binder ratio 0.7

't' = time in years.

Similar formula was proposed by **Kishitani (1960)**[8] in this the author took the effect of the moisture in sand and suggested that a factor of 0.05 be added to the value of water binder ratio to the formula presented by **Uchida and Hamada**

The formula is represented as

$$x = \frac{4.6w - 1.76}{7.2} \sqrt{t}$$
$$x = \frac{w - 0.25}{\sqrt{0.3(1.15 + 3w)}} \sqrt{t}$$

Where

't' is the time in years

'w' is the water binder ratio

The influence of water binder was considered crucial to the carbonation as it affected the permeability of concrete directly; but so, did the reserve alkalinity. Hence **Smolczyk (1969)**[9] proposed a relation between carbonation depth, water binder ratio and compressive strength of concrete mix given in Eq.

$$x = a \left(\frac{w}{\sqrt{N_t}} - b \right) \sqrt{t}$$

Where

'a' and 'b' are coefficients and

'N_t' is compressive strength after 't' days.

A fundamental model was which considers the mass conversation of CO₂, Ca (OH)₂ and C-S-H was been developed by **Papadakis et al. (1989)**[10]. In this model the author considered the products of the higher molar volumes than that of hydroxide and C-S-H after the carbonation. For a fully hydrated concrete the formula for the depth of carbonation is given equation as.

$$x = \left(\sqrt{\frac{2[\text{CO}_2]D_{e,\text{CO}_2}}{[\text{Ca}(\text{OH})_2] + 3[\text{CSH}]}} \right) \sqrt{t}$$

Where

'[CO₂]' is concentration of carbon dioxide in mol/m³

'[Ca (OH)₂]' is concentration of calcium hydroxide in mol/m³

'[CSH]' is concentration of calcium silicate hydrates in mol/m³



' $D_{e,CO}$ ' is diffusivity of carbon dioxide (m²/s)

' t ' is time in seconds

After modelling the above equation with the nineteen different formulas and then incorporating several parameters such as cement content, water-cement ratio, aggregate-cement ratio and humidity, a simplified version of the Eq. for an ordinary Portland cement has been modelled. The formula is presented in Equation.

$$x = 350 \left(\frac{\rho_c}{\rho_w} \right) \frac{\left(\frac{w}{c} \right) - 0.3}{1 + \left(\frac{\rho_c}{\rho_w} \right) \left(\frac{w}{c} \right)} \left(1 - \frac{RH}{100} \right) \sqrt{\left\{ \left[1 + \left(\frac{\rho_c}{\rho_w} \right) \left(\frac{w}{c} \right) + \left(\frac{\rho_c}{\rho_a} \right) \left(\frac{a}{c} \right) \right] y_{CO_2} t \right\}}$$

where ' y_{CO_2} ' is carbon dioxide content by volume in m³

' ρ_c ' is mass density of cement in kg/m³

' ρ_w ' is mass density of water in kg/m³

' ρ_a ' is mass density of aggregate in kg/m³

' w/b ' is water binder ratio

' a/c ' is aggregate-cement ratio

' RH ' is relative humidity in (%)

' t ' is time in years.

2.1 Overview of Mathematical Carbonation model

According to the concrete carbonation model in Papadakis' paper (Papadakis et al., 1991a, 1991b; Papadakis et al., 1989)[11], the carbonation front can be simply predicted by

$$x_c = \alpha_c \sqrt{t}$$

where x_c is the carbonation depth, α_c is the rate coefficient of carbonation, which can be determined

$$\alpha_c = \sqrt{\frac{2C_{CO_2}CO_2]_b}{[Ca(OH)_2]_0 + 3CSH_0 + 3[C_3]_0 + 2[C_2S]_0}}$$

where,

D_{CO_2} is the diffusion coefficient of carbon dioxide in carbonated concrete. $[c_{O_2}]_b$ is the boundary concentration of carbon dioxide, $[Ca(OH)_2]_0$, $[CSH]_0$, $[C_3]_0$ and $[C_2S]_0$ are the initial concentration of carbon dioxide, calcium hydroxide, CSH, C3S and C2S, respectively.

The author **Xiao-Yong Wang** proposed the mathematical equation for the depth of carbonation

follow as [12,13,14]

$$x_c = \sqrt{\frac{2D_c([CO_2]_0/100)t}{0.33CH + 0.214CSH}}$$

$$D_c = A \left(\frac{\varepsilon - \Delta\varepsilon_c}{\frac{C_0}{\rho_c} + \frac{P}{\rho_{HT}} + \frac{W_0}{\rho_w}} \right)^a \left(1 - \frac{RH}{100} \right)^{2.2}$$

$$CSH(t) = 2.85(f_{S,C} * C_0 * \alpha + f_{S,P} * P * \alpha_{HT})$$

where D_c denotes the CO_2 diffusion coefficient, $[CO_2]_0$ denotes the concentration of CO_2 in the external environment, CSH denotes the mass of calcium silicate hydrate, ε is concrete porosity, and $\Delta\varepsilon_c$ denotes the reduction in porosity due to carbonation. $\Delta\varepsilon_c$ can be determined using the variation in the volume of solids between carbonation products and carbonation reactants. RH denotes the relative humidity. A and a are carbonation parameters that can be regressed using the measurement results of the depth of carbonation. shows the calculation of CSH. and fS,P are the mass fractions of SiO₂ in Portland cement and calcined clay, respectively. The terms $f_{S,C} * C_0 * \alpha$ and $f_{S,P} * P * \alpha_{HT}$ denote the mass of CSH from the individual reactions of cement and calcined clay, respectively. The coefficient 2.85 in Eq denotes the molar weight ratio of CSH to SiO₂.

The addition of fly ash increases the carbonation depth of concrete (Thomas, 2013)[15]. Moreover, the carbonation durability should be considered a necessary constraint for the mixture style of fly ash-blended concrete. It implies that the carbon depth of the concrete should be lower than the cover depth. Papadakis et al. (2002) and Papadakis and Tsimas (2002)[16] investigated experimentally and theoretically the carbonation of concrete with supplementary cementing materials. Their studies clarified the effect of the material properties and exposure conditions on carbonation. In addition, they proposed a diffusion-based equation for the carbonation depth of fly ash-blended concrete:

$$x_c = \sqrt{\frac{2D[CO_2]b_0t}{0.218*(C + 0.5*FA)*\alpha_H}}$$

$$D = 6.1*10^{-6} \left(\frac{(V_m - 2)*0.01 + \frac{[W - 0.267*(C + 0.5*FA)*\alpha_H]}{1000}}{\frac{C + 0.5*FA}{\rho_c} + \frac{W}{\rho_w}} \right)^3 \left(1 - \frac{RH}{100} \right)^{2.2} \exp \left[\beta \left(\frac{1}{T_v} - \frac{1}{T} \right) \right],$$

where x_c is the carbonation depth, D the CO_2 diffusivity of concrete, $[c_{02}]_0$ the CO_2 concentration, t the exposure time, and ρ_c and ρ_w are the densities of cement and water, respectively; RH is the relative humidity of exposure, α_H the degree of hydration.

$$\left(\alpha_H = 1 - \exp \left(-3.38 \frac{W}{C + 0.5 * FA} \right) \right)$$

(Oh and Jang, 2004)[17], β the activation energy of CO_2 diffusion ($\beta = 4300$), T_{ref} the reference temperature (293 K), and T the environmental temperature; $(2)*0.01$ V_{air} – represents the increasing concrete porosity due to entrained air (for non-air-entrained concrete, the content of entrapped air is assumed to be 2%) (Czarnecki and Woyciechowski, 2015; Heede et al., 2013; Wong et al., 2011)[18,19,20]. With increasing entrained air volume, the diffusivity of CO_2 and carbonation depth of concrete increase (Czarnecki and Woyciechowski, 2015; Wong et al., 2011)[18,20]. Moreover, the rate of carbonation increases with increasing CO_2 concentration and temperature.



Xiaoetal.[21] obtained a carbonation depth prediction model of RAC by introducing the influence factors of RCA and the 28 groups of experimental data from various countries:

$$D_c = 839g_{RP}g_{RC}(1 - R)^{1.1} \sqrt{\frac{W/(\gamma_c C) - 0.34}{\gamma_{HD}\gamma_c C}} v_0 t$$

$$\gamma_c = 1 - \omega\delta$$

In the formula,

D_c is carbonization depth;

R is relative humidity;

W is water consumption of 1m³ RAC;

C is cement dosage of 1m³ RAC;

γ_c is the cement variety correction coefficient, for Portland cement,

$\gamma_c = 1$, and other varieties of cement,

$\gamma_c = 1 - \beta$, and among of them β is the admixture mass fraction;

γ_{HD} is the correction coefficient of cement hydration degree, when curing exceeds 90d,

$\gamma_{HD} = 1$, when curing is 28d,

$\gamma_{HD} = 0.85$, and the middle curing age is linearly inserted;

n_c is the volume concentration of CO₂;

t is the carbonization time;

g_{RC} is the impact coefficient of RCA, when the substitution rate of RCA is 0%

$g_{RC} = 1$, when the substitution rate of RCA is 0%,

$g_{RC} = 1.5$, and the intermediate substitution rate is based on linear interpolation.

The carbonation prediction model starts from the basic composition of cement, considering the influence of the admixture, and has consistency with the actual application. However, this model fails to consider the effect of RP and the calculation results may be biased. Therefore, a reliable prediction model of carbonation depth is very important for accurate prediction of CO₂ absorption

If completely ignoring the effect of RP in the calculation process of γ_c , it will affect the accuracy of the results. Therefore, the influence coefficient ω of RP on cement hydration is introduced. The revised formula is:

$$D_c = 839g_{RP}g_{RC}(1 - R)^{1.1} \sqrt{\frac{W/(\gamma_c C) - 0.34}{\gamma_{HD}\gamma_c C}} v_0 t$$

$$\gamma_c = 1 - \omega\delta$$

In the formula, g_{RC} is the influence coefficient of RP. When the replacement ratio is 0, $g_{RP} = 1$, when the replacement ratio is 30%, $g_{RP} = 1.075$, and the intermediate substitution ratio is linearly interpolated. ω is the influence coefficient of RP on cement hydration, related to the composition of RP, which is taken 0.3 here; δ is the replacement ratio of RP 3 d, 7 d, 14 d, and 28 d. It can be seen that, when the carbonization depth was small, the obvious measurement error may result in inaccurate results. Considering the error of the test itself, it can be considered that the model can reasonably predict the carbonation depth of recycled concrete with RP.

3.Gaps in the literature review

After an exhaustive literature review, to plug the gaps in the research, the following research needs were identified.

- **Need for a detailed study on how the strength of concrete influences its carbonation resistance**



Even though it is evident in the present scenario that the concrete with least mechanical strength poses a high chance of getting carbonated soon, the relation between the mechanical strength in terms of water binder ratio and the depth of carbonation needs to be examined in detail.

- **Need for the study of influence of mineral admixtures with various water-binder ratios on rate of carbonation**

Though the concept of the durability of concrete with mineral admixtures has been well established, as mentioned in this chapter previously, there have been many contradictions regarding the carbonation resistance of concrete mix with cement partially replaced by mineral admixtures. The optimum dosage of these mineral admixtures, if intended for the use in the concrete mix, in terms of durability should be clarified.

- **Need for an understanding of how porosity affects the ingress of carbon dioxide**

Seldom the concept of porosity and carbonation of concrete has been addressed together. The formation of less porous calcium carbonate after the process of carbonation hinders the further ingress of carbon dioxide into concrete. Hence it is imperative that a study on the porosity of carbonated concrete and rate of carbonation be studied.

- **Need to relate natural and accelerated carbonation and to validate square root t-law of diffusion**

It is still a speculation that the controlled conditions of accelerated carbonation could stimulate the process of long-term natural carbonation. Though research has been done in this area, the phase changes and end products of both natural and accelerated carbonation are yet to be clearly understood. The Fick's law of diffusion (square root t-law) is generally applied to comprehend the rate of carbonation, which assumes steady state diffusion. But as the porosity of carbonated concrete is less than the non-carbonated part, the Fick's law of diffusion should be verified for its validity in carbonation.

- **Need to classify the carbonation resistance of mineral admixture concrete based on fineness**

Most of the literatures mentioned in this chapter conclude that mineral admixture concrete showed less resistance towards carbonation. But there is no dearth of studies that report the effect of fineness of mineral admixtures on increasing the concrete carbonation resistance. A microscopic study on this aspect has not been carried out much; hence to address the phenomenon of fineness of mineral admixtures on carbonation resistance, a detailed study of chemical kinetics is necessary.

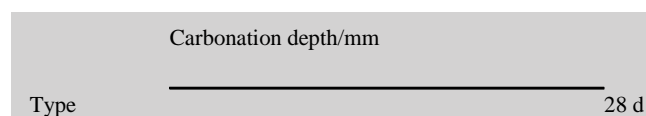
Observations

1. **In the above literature reviews, it was observed that the compressive strength of the specimens gets reduced with the increase in the recycled aggregate.**

Type	C-0-0	C-15-0	C-30-0	C-0-50	C-15-50	C-30-50	C-0-100	C-15-100	C-30-100
7d	28.41	26.52	23.97	25.10	21.91	18.70	21.63	24.37	17.47
28d	37.96	33.01	30.79	30.71	30.32	27.48	29.63	31.54	23.72
90d	38.58	34.47	31.17	30.80	31.38	28.59	29.89	33.22	25.11

Compressive strength of the normal concrete and recycled concrete

2. Carbonation depth of the recycled concrete is greater than of normal concrete at each age of concrete.





	3 d	7 d	14 d	
C-0-0	2.4	3.5	6.3	8.1
C-15-0	2.6	5.5	6.4	9.4
C-30-0	6.5	7.3	8.9	12.0
C-0-50	3.8	6.0	8.2	12.3
C-15-50	5.4	9.1	11.1	15.1
C-30-50	9.7	12.8	17.3	19.8
C-0-100	6.2	10.8	15.1	17.0
C-15-100	8.8	11.2	16.9	21.1
C-30-100	10.5	14.3	19.1	22.3

Carbonization depth of recycled concrete and normal concrete

Here C-15-50 represents Replacement ratio 15% and recycled concrete aggregate replacement ratio 50%

3. At normal days the carbonation depth at respective days 7d,14d,21d was inaccurate the accurate values were observed at 90 days
4. While increasing the entrained air volume the diffusivity of carbon dioxide and carbonation depth of concrete increases.
5. More over the rate of carbonation increases with the increase carbon dioxide concentration and temperature.
6. In addition, with the mineral admixture to the concrete then I was observed that the carbonation depth increased.

Conclusion

1. Based on the above literature reviews in the empirical methods only the few parameters were been considered as also for the accurate results a greater number of parameters were been considered.
2. And considering of the high-volume mass than that of hydroxide and C-S-H and carbonation depth for the full hydrated concrete was done.
3. Carbonation depth of the recycled concrete is greater than of normal concrete at each age of concrete.
4. In empirical studies some gaps are been observed such as the study of the effect of the mineral admixture with various water cement ratio for the carbonation.
5. Among both of the methods are equally useful and important for the carbonation depth prediction but the values of the mathematical approach were found accurate at the at the longer run time and further research should be done to make it more accurate and fast.
6. A combination of these models may provide a more accurate and reliable prediction of carbonation depth in concrete structures.

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