Review on Carbonation Of Concrete Made With Fly Ash And Microsilica and Their Mathematical Models

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Abstract

This paper gives review on different experimental and mathematical models which heavily focus on carbonation of concrete and influence of admixtures like fly ash and micro silica. The article covers the short characteristic of carbonation, its processes and effects. One of the major causes for deterioration and destruction of concrete is carbonation. This paper discusses several issues of uncertainty in prediction and evaluation of carbonation propagation. The paper also heavily focuses and elucidates the gravity of importance, the mechanism of carbonation and how the various parameters like water/cement ratio, admixtures, strength of concrete,and porosity.

The mechanism of carbonation begins with the penetration of carbon dioxide (CO_2) into the concrete porous system to form a corrosive environment by reducing the pH around the reinforcement.

1. Introduction:

Carbonation is widely recognized as a significant cause of corrosion of reinforcement in concrete [1]. Corrosion of reinforcement is found to be the major cause of deterioration of concrete structures all around the world [2, 3]. The basic mechanism involved in this process is that the atmosphere reacts with hydrated cement and destroys its property of alkalinity. The transport properties like porosity, permeability, diffusion and capillarity show their effect on carbonation. They also affect the durability of concrete since they control the influence of chlorides. The paper reviews the effect of curing age on carbonation of concrete made of micro silica and fly ash and it also identifies the relationship between permeability and curing age, strength with the depth of carbonation of concrete. The durability of concrete structures is very much important when the structures are openly exposed to aggressive environments. Carbonation destructs the structure and reduces its service life. Carbonation is a process by which the atmosphere reacts with hydrated cement products to form calcium carbonate thereby alkalinity of the concrete is reduced [4]. In fact, the negative effects of carbonation can be reduced to the maximum extent by using high strength concrete and proper compaction making the concrete denser. The carbonation is found to have a tremendous impact on some of the engineering properties of concrete. Mostly its influence is predominant on compressive strength and hardness than any other properties. The depth of carbonation can be measured by spraying a phenolphthalein indicator on the splitted part of concrete. The solution is a colourless indicator which turns purple when pH is higher than 13.

2. Carbonation process:

Carbonation is a process where carbon dioxide (CO_2) present in the atmosphere reacts in the presence of moisture with the hydrated cement minerals. Carbon dioxide mainly reacts with the calcium hydroxide to form calcium carbonate.

The process of carbonation of concrete involves the following chemical equation which clearly depicts the reaction that takes place between atmospheric CO_2 and the products of cement hydration particularly $Ca(OH)_2$.

$$Ca(OH)_2 + CO_2 \rightarrow CaC0_3 + H_20 \qquad (1)$$

Hydration products (calcium silicate hydrate or CSH gel) and even the residual unhydrated compounds are present in all concrete. They are:

1) Tricalcium silicate (C_3S)
2) Dicalcium silicate (C_2S)

These compounds react with CO_2 as shown in the following reactions:

$$CSH + 3CO_2 \rightarrow 3CaCO_3.2SiO_2 + 3H_2O \qquad (2)$$

$$C_3S + 3CO_2 \rightarrow H_2OSiO_2.H_2O + 3CaCO_3 \qquad (3)$$

$$C_2S + 2CO_2 \rightarrow H_2O\ SiO_2\ H_2O + 2CaCO_3. \qquad (4)$$

Carbonation has two effects, it increases the mechanical strength of concrete, but it also decreases alkalinity, which is essential for corrosion prevention of the reinforcement steel. Carbonation is an unwanted process in concrete chemistry.

3. EFFECTS OF CARBONATION

Some of the beneficial effects of carbonation which can be observed in general are, Carbonated concrete gives an overestimate of strength which in extreme cases can be up to 50 %. It is possible to establish correction factors by removing the carbonated layer and testing the concrete with the rebound hammer on the non-carbonated concrete. A study has been conducted where carbonation is intentionally applied to freshly cast concrete pipes, wherein CO_2 reacts with anhydrous phases to form strength contributing hydrates and carbonates [5]. Because $CaCO_3$ occupies greater volume than $Ca(OH)_2$ which it replaces, the porosity of carbonated concrete is reduced. Also, water released by $Ca(OH)_2$ on carbonation may aid the hydration of unhydrated cement. These changes result in increased surface hardness, increased strength at the surface [6], reduced surface permeability, and reduced moisture movement and increased resistance to those forms of attack which are controlled by permeability. It is well known that carbonation increases the density, strength, modulus of elasticity, and shrinkage of the concrete [7]. Kim [8] et al. studied the effect of carbonation on rebound number and compressive strength of concrete, which has been mentioned by IS 13311 (pt 2): 1992. A strength reduction coefficient is generally applied to the evaluated strength to compensate the effect of surface carbonation.

Detrimental effects of carbonation can be observed as it adversely affects the durability of concrete, as pH of the concrete pore solution is reduced from 12 to less than 9 causing the formation of CaCO3 [9]. This affects the passivation state of embedded steel. When the low pH font reaches the vicinity of the surface of the reinforcing steel, the protective oxide film is removed and corrosion takes place, provided oxygen and moisture necessary for the reactions of corrosion are present [10]. Because of the presence of coarse aggregate, the 'front' does not advance perfectly as a straight line. In many cases, corrosion can take place even when the full carbonation front is still a few millimetres away from the surface of the steel if partial carbonation has taken place[11].

4. Discussion of Experimental Studies For Different Influencing factors

4.1 Effect of water/cement ratio:

From many research studies, it was observed that the water/cement ratio has a profound effect on carbonation irrespective of the mix design [1]. Water/cement ratio was found to be the most authentic and reliable parameter which helps in predicting the resistance of normally vibrated concrete to carbonation [12]. It has been concluded by the earlier research studies that the carbonation depth is directly related to water/cement ratio and it is identified that depth of carbonation increases with increase in water/cement ratio. It is also directly related to the age of the concrete. It has also been found that depth of carbonation has been reduced greatly by reducing water/cement ratio. It is observed at low values of water/cement ratio the depth of carbonation is low but there seems to be a continuous increase in depth of carbonation at higher values of water/cement ratios. Some of the researchers have concluded that carbonation decreases with decrease in water/cement ratio in some tropical arid regions.

4.2. Effect of water curing:

The influence of curing on concrete carbonation can be in terms of the type of curing and in terms of initial curing period. Conclusions drawn upon Impact of curing may also be different for natural long term carbonation with site-curing and accelerated carbonation with laboratory-controlled curing.

In a critical review by Ekolu,et al [44], the author points out that the studies on effects of different curing methods on natural carbonation of concrete in real structures are very rare. N.I. Fattuhi [13] stated the curing periods were increased from 1 to 28 days, the depth of carbonation was reduced to the extent of 61 to 17%. N.I. Fattuhi et al stated that the depth of carbonation decreases with increase of curing period primarily from 1 to 7 days but the change is less significant beyond 7 days. Balayssacetal [14] stated that the depth of carbonation decreases with increase in curing period. But Atis[15] found that carbonation depth has been lower in case of a longer initial curing period. Sulapha et al [16] found that depth of carbonation decreases with increase of curing period. Sisomphon and Franke [17] stated that the depth of carbonation decreased with the increase of curing period in fly ash concrete.

4.3. Effect of type of cement:

Thomas et al. [18] identified that the results of carbonation in fly ash(FA) concrete and ordinary Portland cement (OPC) concrete have been comparable under similar conditions. Alonso et al.[19] concluded that the probability of carbonation in blended cement is higher as compared to OPC. Papadakis [20] concluded that a partial replacement of cement with supplementary cementitious materials (SCMs) results in faster rate of carbonation. Eehab Ahmed Badreldin Khalil Mohamed Anwar [21] observed that the percentage of FA increases the carbonation rate considerably. Duo Zhang et al [22] has stated that FA concrete is more reactive with CO_2 in comparison to OPC concrete. Qingxin Zhao et al [23] and Wang and Park [24] have observed that the average carbonation depth of fly ash(FA) concrete decreases as compared to OPC concrete.

4.4 Effect of Admixtures:

As it was observed from the previous studies strength of concrete reduces with increased depth of carbonation. Many of the industrial by-products like GGBS, SF, FA, Metakaolin etc are increasingly used in constructions as they contribute to the improvement of various other properties [25]. They improve strength of concrete and result in reduction of carbonation depth, replacement of cement also facilitates some benefits like reduction of shrinkage, expansion etc. Some of the studies revealed that lower levels of replacement i.e. 0-30% replacements observed beneficial rafter than higher replacements like beyond 60%.

4.4.1. Effect of Micro Silica:

Micro Silica is very effective in reducing permeability of cement in paste form and in concrete it also reduces porosity [26]. It plays a very significant role in the process of development of high strength and high performance concrete. The concrete made of SF will have very good bond strength and offers resistance to abrasion [27]. The compressive strength will also be improved with the addition SF. The increase in compressive strength is also accompanied by an increase in carbonation depth. When SF reacts with calcium hydroxide the pH value of pore solution is reduced and it indicates the initiation of carbonation process [28]. The previous studies have revealed the fact that beyond 10% replacement of micro silica will show its effect on carbonation and is also responsible for corrosion [29].

4.4.2. Effect of Flyash:

The compressive strength is improved by 30% replacement of FA. It was identified from previous studies the depth of carbonation of High Volume Fly Ash (HVFA) with 90 days curing period is nearly equal to plain concrete. It is true that FA increases carbonation depth, but with HVFA with a curing period 90 days reduces the carbonate depth [30].

The study by Thomas et al. [45], concludes that concretes with up to 30% fly ash carbonated to a similar or slightly greater degree compared with OPC concretes of the same strength grade. However, concrete containing 50% fly ash carbonated at significantly greater rates. This study reports carbonation of fly ash concrete, with particular emphasis on the role of curing. It also observes that in some cases increasing the initial curing period from 1 to 7 days had the effect of reducing carbonation by 50%. Concretes with nominal strength grades M25, M35 and M4.5 and a range of fly ash levels (O-50%) were exposed to various treatments and environments during the first 28 days. After 28 days the concrete cubes were stored either internally or externally (sheltered) and the rate of carbonation was monitored.

Also, Papadakis et al. [20] reported that in the case fly ash is introduced as a fine aggregate replacement, the carbonation rate is reduced. (Khunthongkeaw et al. [46]) However, reaction products between pozzolanic silica and Ca(OH)2 result in the denser structure of the HCP, so diffusivity is reduced, and carbonation is likely to be slowed down.

4.5. Effect of porosity:

Increase in porosity results in increasing depth of carbonation It means that when water content is increased porosity also increases which results in increase of depth of carbonation [26]. When water content is reduced porosity is also reduced this results in reduction of depth of carbonation. Further it was observed that carbonation depth is increased in the environment where CO2 presence is more [31].

In a study by Roy et al. [47] the relationship between carbonation and nature of the pores in the concrete was studied in detail. Larger pores gave rise to higher carbonation depths. Twenty concrete panels were cast and cured for 28 days and kept in two different locations for 2 years. Upon completion of the process, the median pore diameter was determined and found that the median pore diameters for the unmolded samples (with large carbonation depth) are greater than those of the moulded samples (smaller carbonation depth). The result presented which composes the carbonation depth with the median pore size and shows a higher carbonation depth for increased pore size is also expected since a higher porosity will produce a higher diffusion rate for carbon dioxide. The importance of uniform and well compacted concrete is also demonstrated.

4.6. Effect of permeability:

The permeability of hardened cement paste is affected by the type of cement, the water/cement ratio, the degree of hydration (curing) and the compaction of the concrete. Correspondingly the moisture content of the concrete is also a substantial factor. The presence of water is necessary for carbonation. On the other hand, if the pores of concrete are saturated, the diffusion of carbon dioxide is slow[32]. The diffusion rate of CO_2 in water is about four decades lower than in the air. According to Parrot et al[33], the rate of carbonation reaches its maximum at the range of the relative humidity of 50 to 70 %.

5. Discussion On Some Of The Mathematical Model

On the basis of experimental studies in the past along with analytical hypotheses proposed by many authors and researchers, several mathematical models have been developed to predict the carbonation depth of concrete structure. Several early experimental studies on concrete carbonation, for outdoor conditions Shigeyoshi Nagataki and Eun Kyum Kim[36] et al and for controlled indoor conditions, Tuutti et al [37]; Shigeyoshi Nagataki and Eun Kyum Kim[36] et al ; S. Nagataki and H. Ohga [38]; Qiu-Dong [39] hypotheses that depth of concrete carbonation or the location of “carbonation front” is directly proportional to square root of age of concrete. i.e. √t (Papadakis Vayenas, et al. [35],The expression used as given in equation (5).

$$X_c = A\sqrt{t} \qquad (5)$$

Where, □X_c= carbonation depth after time t, t = carbonation exposure duration, and A= empirical constant. In this model, it is to be noted that variable nature of the diffusivity constant of CO_2 was not considered (Houst and Wittmann 2002). In reality, the value of “A” should depend upon the degree of saturation of concrete, the pore structure of concrete, and the carbon dioxide content of the environment.
A more accurate model, based on the microstructure of concrete, which is quite similar to equation (6) was developed by Papadakis Vayenas et al. 1991[35]. This mathematical model yielded a complex nonlinear system of differential equations in space and time and had to be solved numerically for the unknown concentrations of the materials involved (Papadakis Fardis et al. 1991). This model predicted the Equation 2, for giving depth of carbonation.

$$\square = \sqrt{\frac{2\square^{\square}_{\square,\square\square 2}[\square\square_2]^0\square}{[\square\square(\square\square)_2]^0 + 3[\square\square\square]^0 + 3[\square_3\square]^0 + 2[\square_2\square]^0}}$$

(6)

Where $D^C_{e,co2}$, = effective diffusivity of CO_2, $[Ca(OH)_2]^0$, $[CSH]^0$, $[C_3S]^0$ and $[C_2S]^0$ are the "initial" concentrations (at t=0) of $Ca(OH)_2$ (aq), $Ca(OH)_2$ (s), CSH, C_3S, and C_2S respectively at the end of moist curing. In this model, carbonation effects of anhydrous cement clinker phases, concrete porosity and degree of

saturation were also taken into account. The proposed model could predict the test results of previous researchers, for exposure to natural or higher concentrations of ambient CO_2.
This equation was derived with the simplifying assumption that the full carbonation of concrete is not only a complete consumption of calcium hydroxide, but also other mineral phases. However, a very big difference in the rate of carbonation for these phases and Portlandite shows that only the carbonation of $Ca(OH)_2$ is important in practice.
The models were derived and experimentally verified assuming an initial maximum time of concrete early curing, which in practice is not always possible and expedient.

Russell et al. [40] proposed an empirical model defined on the basis of samples prepared in order to achieve a uniform level of humidity in its entire volume:

$$X = \square_0+\square_1[\log(Pl)]+\square_2(f_{cu})+\square_3[RH_{10}]+\varepsilon_1 \qquad (7)$$

where Pl – initial air permeability through the concrete, RH10 – relative humidity in the pores of the concrete toa depth of 10 mm, fcu – compressive strength, βi and εi – coefficients of the model. Meanwhile, Russell et al [40], verifying above findings showed limited compliance of this model with the results obtained for the different concretes – yielding correlation coefficients not exceeding the value of 0.7.

Uomoto et al. [41] presented a complex model of carbonation as a function of w/c, CO2 concentration, temperature and exposure time:

$$\square = (2.084 - 0.8471\square\square\square\square\square_2) \times \square^{8.748-\frac{2563}{\square}} \times [2.39(\frac{\square}{\square})^2 + 0.446(\frac{\square}{\square}) - .398] \times \sqrt{\square_{\square\square 2} \times \square}$$

(8)

where C_{CO2} – CO_2 concentration, %; T – ambient temperature, K; t – time of exposure, years, w/c – water-cement ratio. The model was determined on the basis of research in the natural and accelerated conditions (elevated CO_2 concentrations), but only on samples with a high degree of saturation with water (about 80%), which limits the versatility of the model.
The Bulletin 238 Comite Euro-Internationale du Beton [42], developed by the Working Groups CEB 5.1 Modeling of Material Behavior Exposed is the Environment and the Environmental Actions 5.2, presents a probabilistic model in the form:

$$\square = \square\sqrt{2\square_1\square_2\square_3\Delta\square}\sqrt{\frac{\square_{\square\square\square}}{\square}\square} \times (\frac{\square_{\square}}{\square})^{\square}$$

(9)

where D_{nom} – diffusion coefficient at RH =~ 65% and text =~ 20^0C, mm^2 /year; Δc – difference of CO_2 concentration on the surface of concrete and inside the concrete; the authors suggest to assume the value of concentration equal to 0 at the depth of carbonation range, and then Δc in Eq. (5) is equal to C_0 – concentration of CO_2 in the air, kg/m^3 ; a – amount of CO_2 for concrete full carbonation, calculated with Eq. (6) kg/m^3

$$a = 0.75 \cdot C \cdot [CaO] \cdot \alpha H \cdot (M_{CO2}/M_{CaO}) \qquad (6)$$

wherein the C – content of cement, kg/m3 ; [CaO] – CaO content in the cement composition; αH – the degree of hydration of cement; M_{CO2} and M_{CaO} – molar masses; n – coefficient of influence of changes in external conditions; fro laboratory conditions n = 0, for natural conditions n ≤ 0.3 (the higher the changes of conditions

the higher value of n); t_0 – reference period – in natural conditions t0 = 1 climatic year; t – time of exposition, years.
k_1 – coefficient of concrete humidity, from 0.3 to 1.0; k_2 – coefficient of time of early curing; 1.0 for "good" curing or 2.0 for "bad" curing; there is no definition of "good" and "bad" curing; k_3 – coefficient of concrete bleeding, equal from 1.0 (minimum of bleeding) to 1.5 (for high degree of bleeding)

Jiang et al. [43] presented the model in the form of equation:

$$\square = 839(1 - \square\square)^{1.1} \times \sqrt{\frac{\frac{\square}{\square_{\square}\square} - 0.34}{\square_{\square\square}\square_{\square}\square}} \times \square \times \sqrt{\square} \qquad (10)$$

where RH – relative humidity, w and c – cement and water content in concrete, ϕ – CO2 concentration, rc – coefficient of type of cement, rHD – degree of cement hydration. In further studies, the same authors [42] modified model (7) into a form suitable for so-called high volume fly ash concrete (HVFAC):

$$\square = 839(1 - \square\square)^{1.1} \times \sqrt{\frac{\left(\frac{\square}{\square}\right)^{*} - 0.34}{\square_{\square}(1-\square\square)\square}} \times \square \times \sqrt[\square]{\square} \qquad (11)$$

where αH – the degree of hydration of cement, k – coefficient taking into account the mass content of reactive pozzolans in the ashes, β – the degree of hydration of ashes, (W/B) ∗ – efficient water-binder ratio, taking into account the reactivity of the ashes and its maximum amount that can react due to the pozzolanic mineralogical composition of the ash and cement.

5. Experimental Methods involved in the determination of carbonation

5.1. Fourier Transform Infrared (FTIR) Spectroscopy

Fourier transform infrared (FTIR) spectroscopy is a measurement technique that allows one to record infrared spectra. Infrared light is guided through an interferometer and then through the sample (or vice versa). A moving mirror inside the apparatus alters the distribution of infrared light that passes through the interferometer. The signal directly recorded, called an "interferogram", represents light output as a function of mirror position. A data processing technique called Fourier transform turns this raw data into the desired result (the sample's spectrum). The IR spectrum of each powder sample mixed with KBr in a proportion of 1:10 was obtained to facilitate quantitative measurement of the depth of carbonation. The background spectrum of the laboratory environment was scanned before the powder sample was scanned. Carbonation is represented by the transformation of the CjO bonds of CO2 into C–O bonds in the $CaCO_3$. Thus, a study of the characteristic peak of the C–O functional group in the wave number range of 1410–1510 cm would identify the carbonation in concrete.

5.2. X-Ray Diffraction (XRD)

X-ray diffraction is a tool used for identifying the atomic and molecular structure of a crystal, in which the crystalline atoms cause a beam of incident X-rays to diffract into many specific directions. By measuring the angles and intensities of these diffracted beams, a crystallographer can produce a three-dimensional picture of the density of electrons within the crystal. From this electron density, the mean positions of the atoms in the crystal can be determined, as well as their chemical bonds, their disorder and various other information. XRD analysis was conducted on a Shimadzu Model XD-3. The relative diffraction peaks of $CaCO_3$ and $Ca(OH)_2$ in powder X-ray diffraction analytical spectra of concrete powder dried at 105^0 C were compared using an apparatus for powder X-ray diffraction analysis. Its operating conditions were CuKa radiation (40 kV, 100 mA); scan rate: 48/ min; 2θ: 10–700 . It is evident that there exists a peak for $CaCO_3$ but no peak for $Ca(OH)_2$; hence, this sample could be regarded as fully carbonated. There exists only a peak for $Ca(OH)_2$ and no peak for $CaCO_3$; hence, the sample could be regarded as being non carbonated. Both peaks $CaCO_3$ and $Ca(OH)_2$ exist , which indicates that the sample is partly carbonated.

XRDA and FTIR test methods were used to determine the true depth of carbonation in concrete. Each test method has its advantages, but the resulting depths of carbonation are similar[34].

5.3. Test Of Depth Of Colorless Region Using Phenolphthalein Indicator

The majority of research works on concrete carbonation use a phenolphthalein indicator to assess carbonation depth. This involves spraying concrete broken faces after flexural strength tests with 1% phenolphthalein in 70% ethyl alcohol.

1. When the pH of the pore solution is less than 7.5, the degree of carbonation of the specimen is 100%.
2. The pH value of the pore solution is between 7.5 and 9.0, the degree of carbonation is 50– 100%.
3. pH of the pore solution is 9.0–11.5, the degree of carbonation is 0–50%.
4. pH of the pore solution exceeds 11.5, the specimen is not carbonated.

6. Conclusions:

General conclusions are drawn from the extensive review of literature on carbonation on concrete:

1. Many years of research conducted by the authors presented synthetically in this article proved that the process of carbonation, although infinite in time is limited in the space of concrete, as a result of self-limiting nature of the phenomenon (gradual lowering of the CO2 diffusion ratio and the rate of carbonation tending asymptotically to zero).

2.The influence of water/cement ratio is very much significant on the strength of the concrete since the strength of the concrete depends on water/cement ratio. The depth of carbonation depends on water/cement ratio higher water/cement ratio contributes to higher carbonation depth.

3. Increase in curing period reduces the carbonation depth. The resistant power of the concrete to carbonation is increased with sufficient and substantial curing periods.

4. The compressive strength also depends on water/binder ratio and mineral admixtures both higher and lower water/binder ratio affects the properties of concrete.

5. Higher porosity gives rise to higher carbonation depths. To ensure lower porosity or denser structure, concrete should be uniform, well compacted and has high cement content. Carbonation causes a reduction in porosity of the concrete. Therefore, carbonation slows down with time or its propagation. Pore blocking due to rain may also influence carbonation propagation. Hence, in a porosity model, a function which incorporates initial concrete properties as well as other variables like degree of carbonation, relative humidity, temperature etc. must be used.

6. Equation 5, which defines a linear relationship between carbonation depth and the square root of time with a proportionality constant, has been used by several researchers for reporting the effects of influencing parameters on carbonation of concrete. This equation considers diffusion properties to be constant which does not reflect the real scenario. Also, as observed across different experimental data the proportionality-constant changes appreciably. Although highly convenient in comparing results in controlled laboratory conditions, this equation should not be used to study or predict long-term natural carbonation of concrete.

7. In general, the carbonation depth decreases as aggregate replacement by SCM increases and increases as cement replacement by SCM increases. Concretes with up to 30% fly ash carbonated to a similar or slightly greater degree.

8. As the humidity level increases from around 50% to 75%, there is a significant increase in carbonation depth with increasing humidity level. There is then a decrease in carbonation depth as the relative humidity increases from 75 to 84%.

6. References:

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