# Numerical study in the influence of carbonation on the penetration of chloride into concrete

C. Y. Kim Samsung Engineering, Seoul, Korea

J. K. Kim

Korea Advanced Institute of Science and Technology, Daejeon, Korea

ABSTRACT: For a reasonable description of combined deterioration – that is, the chloride attack accompanying carbonation, an integrated numerical analysis program, which can simulate the chloride penetration, moisture transport, and carbonation in the same time and space domain, is developed in this study. It is assumed that the main reason for the accelerated penetration of chloride under combined deterioration is re-dissolution of bound chloride as a result of carbonation. After verifying the applicability of developed analysis program by performing a simulation analysis of Lee & Yoon's experimental work, case studies were made in order to investigate the influence of carbonation on the penetration of free chloride under various environmental conditions.

# **1** INTRODUCTION

As commonly known, chloride attack and carbonation are the main reasons for the initiation of steel corrosion in concrete, and the mechanisms and influencing factors for each phenomenon are clearly defined owing to the numerous research efforts. Although recent experimental observations imply the strong interaction between carbonation and chloride attack, these aspects have been separately considered in most of the existing researches on the estimation of time for the initiation of steel corrosion, which is commonly recognized as service life of RC structures. Considering the facts that RC structures exposed to natural environment are basically under combined deterioration - carbonation and chloride attack, and the several experimental results show that the penetration of chloride ion is more pronounced when concrete is exposed to combined deterioration, separate consideration of these aspects can cause an unfavorable overestimation of service life. The purpose of this study is to develop a numerical analysis procedure which can reasonably describe the penetration profiles of chloride in concrete exposed to combined deterioration. It is assumed that the main reason for the accelerated penetration of chloride under combined deterioration is redissolution of bound chloride as a result of carbonation. Since both chloride penetration and carbonation are closely related to humidity distribution in concrete, humidity analysis is also integrated in the developed analysis process and all the aspects are analyzed in the same time and space domain.

# 2 ANALYSIS MODELS

# 2.1 Chloride penetration

## 2.1.1 Diffusivity of chloride ion

The model equation adopted by ACI is selected for the numerical analysis, as it can consider most of the influencing factors (Thomas & Bentz 2000). It should be noted that the model equation for the influence of moisture content is taken from Saetta's, as that factor is not included in ACI model equation (Saetta et al. 1993). The model equation is expressed as follows,

$$D_{\rm Cl}(w/b,t,T,RH) = f_{\rm Cl,1}(w/b)f_{\rm Cl,2}(t)f_{\rm Cl,3}(T)f_{\rm Cl,4}(RH)$$
(1)

where  $D_{Cl}$  is diffusivity of chloride ion (m<sup>2</sup>/s), w/b is water/binder ratio, t is age (day), T is absolute temperature (K), RH is relative humidity (%), and  $f_{Cl,i}$ represent the correction functions for each influencing factor (*i*=1: water/binder ratio, *i*=2: age, *i*=3: temperature, *i*=4: relative humidity) (Kim 2008).

## 2.1.2 Binding of chloride ion

Freundlich isotherm is selected for the development of analysis program.

$$C_b = g C_f^{\ h} \tag{2}$$

For the constants in Equation (2), the following model equations derived from Glass et al.'s experimental results are adopted (Glass et al. 1997, Han 2007).

$$g = C(0.056 + 0.025C_3A)/100$$
 (3a)

$$h = \frac{1}{0.076C_3A + 1.91} \tag{3b}$$

where C is unit cement content (kg/m<sup>3</sup> concrete), and C<sub>3</sub>A is tricalcium – aluminate (C<sub>3</sub>A) content of cement (%).

## 2.2 Carbonation

#### 2.2.1 Rate of carbonation

For the rate of carbonation reaction, the following approximate equation is adopted (Danchkwerts 1970, Papadakis et al. 1991).

$$r_{\rm CH} = -\frac{\partial [{\rm Ca(OH)}_2]}{\partial t}$$
  
=  $HRTk_2[{\rm OH}^-]_{eq}[{\rm CO}_2]f_{r{\rm CH},1}(RH)f_{r{\rm CH},2}([{\rm Ca(OH)}_2])$   
(4)

where  $[OH^-]_{eq}$  is molar concentration of hydroxyl ion at equilibrium condition (i.e. at saturation; = 43.2 mol/m<sup>3</sup>),  $k_2$  is rate constant for the reaction (= 8.3 m<sup>3</sup>/(mol sec)), *H* is Henry's constant for the dissolution of CO<sub>2</sub> in water (= 34.2 mol/(m<sup>3</sup> atm) at 25 °C),  $f_{rCH,1}$  and  $f_{rCH,2}$  are correction factors depending on the relative humidity and the concentration of Ca(OH)<sub>2</sub> (Saetta & Vitaliani 2004, 2005).

#### 2.2.2 Diffusivity of carbon dioxide

The following equation, taken partly from Papadakis and Saetta's research work, is selected for the carbonation analysis (Papadakis et al. 1991, Saetta & Vitaliani 2004, 2005).

$$D_{\rm CO_2}(p_c, RH, T) = f_{\rm CO_2, 1}(p_c) f_{\rm CO_2, 2}(RH) f_{\rm CO_2, 3}(T)$$
(5)

where  $D_{\text{CO2}}$  is diffusivity of CO<sub>2</sub> (m<sup>2</sup>/s),  $p_c$  is porosity of hardened cement paste, and  $f_{\text{CO2},i}$  are correction factors for each influencing factor (*i*=1: porosity, *i*=2: relative humidity, *i*=3: temperature) (Kim 2008).

#### 2.3 Moisture transport

#### 2.3.1 Diffusivity of humidity

For the humidity analysis, the fundamental part of the diffusivity is taken from the popular equation introduced in CEB-FIP model code, which is a function of the strength and relative humidity of concrete, and the influence of temperature is considered by using the equation proposed by Mihashi (Mihashi & Numao 1989). The model equation adopted for the humidity analysis is as follows:

$$D_{w}(f_{ck}, RH, T) = D_{w,1}(f_{ck})f_{w,1}(RH)f_{w,2}(T)f_{w,3}(T)$$
(6)

where  $D_w$  is diffusivity of humidity (m<sup>2</sup>/s),  $f_{ck}$  is characteristic compressive strength of concrete (MPa),  $D_{w,1}$  is the diffusivity of humidity when the relative humidity equals 100 % (m<sup>2</sup>/s), and  $f_{w,i}$  are correction factors for each influencing factor (*i*=1: relative humidity, *i*=2, 3: temperature) (Kim 2008).

#### 2.3.2 Sorption isotherm

It is assumed that the adsorption and desorption isotherms are the same; this might be an acceptable approximation for the environment where concrete is exposed to cyclic wetting and drying. The equation for the sorption isotherm is derived on the basis of the model equations proposed by Bazant, which originally were separately defined for the adsorption and the desorption (Bazant & Baweja 1995, Xi et al. 1994). The adopted equation is as follows:

$$S = 0.2 + 0.8 \left(\frac{RH}{98}\right)^3$$
(7)

where S is degree of pore saturation in concrete. Analysis procedure

### 2.4 Governing equation

#### 2.4.1 Moisture transport analysis

Assuming that the moisture transport in concrete is isotropic and the self desiccation due to hydration of concrete is negligible, the governing equation of moisture transport, which describes the time dependent change of moisture distribution in concrete, is expressed as follows:

$$\frac{\partial(RH)}{\partial t} = D_{w} \nabla^{2}(RH)$$
(8)

where  $\bigtriangledown^2$  is Laplacian operator.

#### 2.4.2 *Carbonation analysis*

Carbonation process is composed of two aspects, penetration of  $CO_2$  and carbonation reaction. Based on the diffusion theory and the mass conservation considering the consumption of  $CO_2$  due to carbonation reaction, the governing equation for the penetration of  $CO_2$  can be derived as follows:

$$\frac{\partial}{\partial t} \left[ p(1-S)[\mathrm{CO}_2] \right] = \nabla \Box (D_{\mathrm{CO}_2} \nabla [\mathrm{CO}_2]) - pSr_{\mathrm{CH}} \qquad (9)$$

The degree of saturation, *S* can be calculated from the result of humidity analysis and the sorption isotherm. By substituting Equation (4) for  $r_{\text{CH}}$  in Equation (9), we can get the following equation:

$$\frac{\partial [\mathrm{CO}_2]}{\partial t} = \frac{D_{\mathrm{CO}_2}}{C_1} \nabla^2 [\mathrm{CO}_2] - C_2 [\mathrm{CO}_2]$$
(10)

where 
$$C_1 = p(1-S)$$
  
 $C_2 = \frac{pS \times HRTk_2[OH^-]_{eq} f_{rCH,1} f_{rCH,2}}{C_1}$ 

As carbonation progresses, the remaining amount of  $Ca(OH)_2$  in concrete at time t (=[ $Ca(OH)_2$ ](x, y, z, t)) is gradually decreasing. The consumption rate of Ca(OH)<sub>2</sub> is expressed as follows:

$$\frac{\partial}{\partial t} [Ca(OH)_2] = -pSr_{CH}$$
(11)

Therefore,  $[Ca(OH)_2](x, y, z, t)$  can be calculated as follows:

$$[Ca(OH)_{2}](x, y, z, t) = [Ca(OH)_{2}]_{0} - \int_{\tau=0}^{\tau-1} pSr_{CH}d\tau$$
(12)

where  $[Ca(OH)_2]_0$  is the initial amount of  $Ca(OH)_2$ available for carbonation (Papadakis et al. 1991). In the carbonation analysis, carbonation depth is determined on the basis of remaining amount of  $Ca(OH)_2$ . Although exact value of  $[Ca(OH)_2]$  for the determination of carbonation front has not been clearly defined yet, it is assumed that concrete is carbonated if 90% of  $[Ca(OH)_2]_0$  are consumed by carbonation reaction. This assumption is fairly consistent with Houst & Wittmann's experimental observation (Houst & Wittmann 2002).

#### 2.4.3 *Chloride attack analysis*

Based on the diffusion theory and the mass conservation considering the binding of chloride ion and the convective flow of chloride ion, the governing equation for the penetration of chloride ion can be derived as follows:

$$\left(\frac{\partial C_b}{\partial C_f} + pS\right)\frac{\partial C_f}{\partial t} = \nabla [[pSD_{cl}\nabla C_f] + pC_f\frac{\partial S}{\partial t}$$
(13)

The second term in the right side represents the convective flow of chloride ion due to moisture transport. The rate of moisture change  $(=\partial S/\partial t)$  can be calculated from the result of humidity analysis and the sorption isotherm. The binding capacity term  $(=\partial C_b/\partial C_f)$  in the left side is approximately estimated from the previously calculated concentration of free chloride  $(=C_t^{t-\Delta t}: C_f)$  at time  $t-\Delta t$ .

Appling Equation (2) to Equation (13) yields the following equation:

$$\frac{\partial C_f}{\partial t} = \frac{pSD_{Cl}}{C_3} \nabla^2 C_f + C_4 C_f$$
(14)

where 
$$C_3 = \left( gh \left( C_f^{t-\Delta t} \right)^{h-1} + pS \right)$$
  
 $C_4 = \frac{p}{C_3} \frac{\partial S}{\partial t}$ 

#### 2.5 Integrated analysis procedure

If concrete is exposed to carbonation and chloride attack simultaneously, penetration of chloride ion cannot be readily described on the basis of diffusion theory. The concentration profile of chloride ion for combined deterioration shows very different behavior from that for single deterioration, i.e. chloride attack only. Several experimental results show that the penetration of chloride ion is more pronounced when concrete is exposed to carbonation as well as chloride attack (Lee & Yoon 2003, Oh et al. 2002, 2003, 2005, Song et al. 2006). It is commonly recognized that the main reason for this phenomenon is redissolution of bound chlorides as a result of pH drop in pore solution due to carbonation. To consider this aspect, it is supposed that the coefficient g in Equation (2) is gradually decreasing as carbonation progresses; i.e. g is expressed as a function of the concentration of CO<sub>2</sub>. Although Glass states that most of bound chlorides are released due to carbonation (Glass et al. 2000), it is assumed that part of bound chlorides still remains fixed to concrete even after concrete is fully carbonated.

Incidentally, a number of observations indicate that the concentration of chloride ion at the surface of concrete increases with exposing time, even for concrete under submerged condition (Tang & Gulikers 2007, Tang & Nilsson 2000, Thomas & Bentz 2000). Following the Tang's opinion, this aspect is modeled by changing the binding capacity, i.e. g, with time; exponential function is used for that purpose (Kassir & Ghosn 2002).

Consequently, modified g, denoted as  $g_m$ , is expressed as follows:

$$g_m([CO_2], t) = g \times f_{g,l}([CO_2]) f_{g,2}(t)$$
 (15)

Correction factors in Equation (15) are defined as follows:

$$f_{g,1}([CO_2]) = rc_{\min} + (1 - rc_{\min}) \left(\frac{[CO_2]_s - [CO_2]}{[CO_2]_s}\right)^{n_c}$$
(16a)

where  $[CO_2]_s$  is CO<sub>2</sub> concentration at the surface of concrete,  $rc_{min}$  is the relative amount of bound chlorides when the concentration of CO<sub>2</sub> is the same with  $[CO_2]_s$ , i.e. minimum value for relative amount of bound chlorides, and  $n_c$  is a constant.

$$f_{g,2}(t) = 1 - e^{-k_b t}$$
(16b)

where  $k_b$  is a constant which determines the increasing rate of binding capacity (year<sup>-1</sup>).

In order to apply Equation (15) to chloride attack analysis, governing equation of chloride penetration, i.e. Equation (14), should also be modified accordingly. Figure 1 describes the situation when binding isotherm is changing with time, from t- $\Delta t$  to t. Assume that the condition for concentration of chloride ion is initially represented by point A. If we use  $(\partial C_b/\partial C_f) \times \partial C_f$  to calculate the change in concentration of bound chlorides (= $\partial C_b$ ), we only get the  $C_b$ value for point A'. The actual point representing the current condition, i.e. condition at time t, is B'. The length between A' and B' must be subtracted from  $C_b$  value for point A'. If we assume that the length between A' and B' is the same with that between A and B, we can obtain the following governing equation:

$$\frac{\partial C_f}{\partial t} = \frac{pSD_{Cl}}{C_3} \nabla^2 C_f + C_4 C_f + \frac{C_5}{C_3 \Delta t}$$
(17)

where  $C_5$  is the length between A and B



Figure 1. Change in binding isotherm.

In reality, moisture transport, carbonation, and chloride attack are closely connected to each other; each aspect is dependent on the other aspects. For the accurate description of real phenomenon, these aspects should be analyzed in the same time and space domain. Although the coupling effect among these aspects can best be investigated by means of iteration method, each aspect is analyzed separately and successively, from the moisture transport to chloride attack, in order to simplify the analysis procedure and save the calculation time. Analysis priority for each aspect is determined from its influence to the other aspects, thereby the analysis error due to avoiding iteration can be minimized. For instance, greatest priority is given to moisture transport analysis because it affects all the other aspects.

#### **3 VERIFICATION ANALYSIS**

Despite its importance on the accurate estimation of service life of RC structures, only limited researches have been performed on experimental investigation or analytical description of the combined deterioration (Jang 2003, Kwon 2006, Kwon et al. 2007, Lee & Yoon 2003, Oh et al. 2002, 2003, 2005, Song et al. 2006). Among those, Lee & Yoon's experimental results are selected for the verification of developed analysis program because the concentration profile of total chlorides are clearly arranged according to the exposing times and water/cement ratios of concrete in their reported data. In this experiment, concrete specimens are exposed to cyclic wetting and drying environment which simulates combined deterioration; 1-week wetting by submerging in 0.5M NaCl<sub>2</sub> solution (chloride attack) followed by 1-week drying by exposing to air where relative humidity is 65% and concentration of  $CO_2$  is 5% by volume (carbonation).

Before performing a combined deterioration analysis, diffusivities of carbon dioxide for each concrete mixture were determined from the reported carbonation depth history (Kim 2008). After then, the diffusivities and binding isotherms of chloride, and constants related to re-dissolution of bound chloride, which can best reproduce the experimental data, are subsequently determined. Manually determined material parameters for each mixture proportion are summarized in Table 1. The other material parameters, which don't appear in Table 1, are directly calculated from the model equations introduced in Chapter 2. Additionally, instead of using Equation (16b), it is assumed that  $f_{g,2}(t) = t/t_{ref}$  where  $t_{ref}$  is the reference time (= 1 year), i.e. binding capacity increases in proportion to the exposing time. Although actual

Table 1. Manually determined material parameters for the verification of combined deterioration.

w/c	$f_{{ m CO2,1}} \ (10^{-8}  imes { m m}^2/{ m s})$	$f_{{ m Cl},1} \ (10^{-12}  imes { m m}^2/{ m s})$	g	h	rc <sub>min</sub>	n <sub>c</sub>
0.45	7.95	3.35	1.823	0.35	0.425	1.0
0.50	25.5	6.21	2.057	0.35	0.374	1.0
0.55	64.3	10.9	2.288	0.35	0.383	1.0

value of g might be underestimated by this assumption, this is a reasonable approximation for the early age concrete – i.e. before the age of 1 year, because usual value of  $k_b$  in Equation (16b) is 0.25 (Kassir & Ghosn 2002).

Figure  $2(a) \sim 2(c)$  describe the calculated concentration profiles of total chlorides as well as those measured experimentally. The measured concentration profiles of total chlorides seem very abnormal; maximum concentration is observed about  $3 \sim 15$  mm inside the surface of concrete even though the chlorides are supplied from the surface. This strange phenomenon cannot be described solely by the diffusion theory. A number of researchers found out that this abnormal behavior can be properly reproduced if binding capacity of concrete is modeled as a function of carbonation degree (Jang 2003, Kwon 2006, Kwon et al. 2007, Song et al. 2006). As mentioned in Chapter 3.2, a similar idea on the basis of the concentration of CO<sub>2</sub> rather than the degree of carbonation is adapted in this study. Since analysis results in Figure  $2(a) \sim 2(c)$  show fairly good agreement with the experimental data, it can be said that the assumptions made and the analysis algorithm developed in this study can explain the real nature of combined deterioration quite reasonably. According to the analysis result, the point where maximum concentration of total chlorides is observed corresponds with the CO<sub>2</sub> penetration front – the nearest point from the surface where the concentration of  $CO_2$  is zero; usually, the depth measured from the surface of concrete to this point is about  $3 \sim 5$  mm deeper than the carbonation depth. Figure 3(a) and 3(b) display the calculated concentration profiles of free chlorides and bound chlorides for w/c=0.5, respectively. Due to the influence of convective flow and the moisture distribution, the concentration of free chlorides at the surface is slowly increasing with the exposing time. After 6 months, however, the rate of increase in the concentration of free chlorides becomes negligible. On the contrary, since it is assumed that the bounding





Figure 2. Verification of combined deterioration analysis.



Figure 3. Concentration profiles of chlorides for combined deterioration (w/c=0.5).

capacity increases with the exposing time, the concentration of bound chlorides is continuously increasing. It means that the increase in concentration of total chlorides at old age is mostly attributed to the time dependent change in binding capacity. As this observation is derived solely from the numerical analysis, it needs to be verified by further experimental researches.

## 4 CASE STUDY

As mentioned before, for RC structures exposed to combined deterioration, the penetration depth can be extended because of the re-dissolution of bound chlorides due to carbonation. Therefore, the penetration of free chlorides can be different according to the carbonation depth even if the supplied amount of chlorides is the same. To investigate the influence of carbonation depth on the increase in the penetration depth of free chloride, a case study is performed about two kinds of environment - cyclic wetting and drying, wetting and permanent drying – for various levels of surface CO<sub>2</sub> concentration. In the cyclic wetting and drying condition, concrete is exposed to 1-week drying after 1-week wetting periodically, while concrete is exposed to drying condition permanently after 10-weeks of wetting in the wetting and permanent drying condition. Four different levels of  $CO_2$  concentration – 0.5, 1, 3, and 5% vol. air, are selected to manually change the carbonation depth. Water/cement ratio is assumed to be 0.5. The other material properties, e.g. surface concentration of chlorides, are the same as those used in the verification analysis. Time dependent change in the binding capacity is not considered, i.e. it is assumed that  $k_b = \infty$ . Other parameters related to binding isotherm are assumed as follows:  $rc_{\min} = 0.4$ ,  $n_c = 1.0$ .

Figure 4 shows the concentration profiles of chloride for cyclic wetting and drying condition after 1 year of exposure. As depicted in the figure, the penetration depth of free chloride is slightly increased as carbonation depth increases. However, the increase is not so apparent because most of the re-dissolved chlorides during the drying period are escaped from the concrete during the following wetting period. On the contrary, for the wetting and permanent drying condition, chlorides penetrated into concrete during the wetting period can never escape from the concrete again. Therefore, all the redissolved chlorides must move into the concrete, thereby the penetration of free chlorides is accelerated with the increase of carbonation depth. This aspect is clearly shown in Figure 5(a), though the penetration depth of total chlorides is not obviously increased (see Fig. 5(c)). It can be shown in Figure 5(c) that the total amount of chlorides penetrated into concrete remains constant regardless of exposing time. It is obvious because chlorides are supplied during the wetting period only; i.e. total amount of chlorides in concrete cannot be changed during the subsequent drying period. From the result of the case study, it can be concluded that the penetration of free chlorides are increased due to carbonation only when most of the re-dissolved chlorides are able to penetrate into concrete. In other words, carbonation has strong influence on the penetration depth of free chlorides for RC structures located at the splash zone or for bridge decks exposed to deicing salts, but it is not important for the RC structures located at the tidal zone since the re-dissolved chlorides can be washed out during the flowing tide.



Figure 4. Combined deterioration under cyclic wetting and drying. (concentration profiles after 1 year.)

Additionally, another case study representing more realistic problem in situ is performed to investigate the influence of carbonation depth on the increase in the penetration depth of free chlorides. It is assumed that RC structure is exposed to drying condition for 9 months, after exposed to wetting condition for 3months, annually (see Fig. 6(a)). To manually control the influence of carbonation, two different concentration levels of CO<sub>2</sub>, 0.045 (Case I) and 0 % (Case II, no carbonation), is imposed during the drving period. The other input material properties are the same as those used in the verification analysis. Figure 6(b) shows the history of free chloride penetration for Case I and Case II. Penetration depth of free chloride displayed in the figure corresponds to the free chloride concentration of 5 kg/m<sup>3</sup> solution. The history of carbonation depth for Case I is also displayed. It is clearly shown in the graph that the penetration depth of free chloride is extended as carbonation depth increases; increase in free chloride penetration is about  $26 \sim 50\%$  of carbonation depth. Consequently, for the accurate estimation of service life of RC structures located at the splash zone or that of bridge decks, the effect of carbonation should be considered in the chloride attack analysis, or service life could be undesirably overestimated.





Figure 5. Combined deterioration under wetting and permanent drying. (concentration profiles after 1 year)







Figure 6. Case study for the influence of combined deterioration.

## 5 CONCLUSIONS

An integrated numerical analysis program which can reasonably describe the penetration profiles of chloride in concrete exposed to combined deterioration, i.e. chloride attack accompanying carbonation, is developed in this study. Re-dissolution of bound chloride as a result carbonation is considered by using a model equation in which the binding capacity of concrete is expressed as a function of the concentration of  $CO_2$ . The applicability of developed program is investigated by carrying out a verification analysis simulating Lee & Yoon's experimental work, and the result indicates that the abnormal distribution of total chlorides in concrete exposed to combined deterioration can be reasonably described by the developed program.

Case study for the influence of carbonation on the chloride penetration reveals that the increase in penetration depth of free chloride due to carbonation is dependent on the exposed environment; carbonation can accelerate the penetration of free chlorides only when most of the re-dissolved bound chlorides are available for the further penetration into concrete. Another case study simulating more realistic situation clearly shows that the penetration depth of free chloride can be increased by up to 50 % of carbonation depth because of the combined deterioration.

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