

# An Analytical Study on the Carbonation Progress of Concrete Depending on the Quality of the Surface Layer

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## ABSTRACT

This paper theoretically examines and proposes simple computational methods of predicting the carbonation progress of concrete when the qualities of the surface layer and internal concrete are not the same but cementitious materials have been combined at the surface layer of concrete. It is based on the model that carbon dioxide moves at a steady rate of diffusion according to Fick's law; reacts instantaneously with calcium hydroxide ( $\text{Ca}(\text{OH})_2$ ) at the boundary between carbonated and noncarbonated regions; and the boundary moves inward sequentially. The carbonation progress is simulated using data from previous papers.

## KEYWORDS

Quality of the surface layer of concrete, Carbon dioxide diffusion, Simulation of carbonation progress.

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## 1 INTRODUCTION

In a thermo-hygrostatic environment with a constant CO<sub>2</sub> concentration, the carbonation of concrete having a homogeneous quality from the surfaces inward is known to progress in proportion to the square root of time, following the so-called rule of  $\sqrt{t}$  [Japan Institute of Construction Engineering 1986]. This is theoretically explained by the solution of a model expressing a phenomenon in which CO<sub>2</sub> steadily diffuses through a carbonated region of concrete and instantaneously reacts with Ca(OH)<sub>2</sub> at the boundary between the carbonated and noncarbonated regions, thereby progressively shifting the boundary deeper inward [Architectural Institute of Japan 2004]. This hypothesis has also been virtually validated by accelerated tests under thermo-hygrostatic conditions with a constant CO<sub>2</sub> concentration. Moreover, it has been proven to be practically valid by analyses based on unsteady diffusion [Maeda 1989, Fukushima 1991 & Masuda et al. 1991]. The progress of carbonation can therefore be expressed by Eq. (1).

$$C = A\sqrt{t} \quad (1)$$

where  $C$  : carbonation depth(mm)

$t$  : time(years)

$A$  : coefficient of carbonation speed(mm/year<sup>1/2</sup>)

The coefficient of carbonation speed may also be expressed by Eq. (2) as a coefficient determined by the effects of various factors [Architectural Institute of Japan 2004].

$$A = \alpha\beta\gamma\delta \quad (2)$$

where  $\alpha$  : coefficient for environmental conditions (CO<sub>2</sub> concentration, temperature, humidity, etc.)

$\beta$  : coefficient for concrete qualities (W/C, strength, cement type, etc.)

$\gamma$  : coefficient for defects related to placing (cracking, degree of honeycombs, etc.)

$\delta$  : coefficient for finishing material (gas permeation resistance, type, etc., of finishing material)

The quality of the surface region of architectural concrete in actual buildings is said to be rougher than that of internal concrete due to the effect of drying [Ohta Tatsumi et al. 2003 & Yuasa et al. 1998]. The quality of concrete is also made inhomogeneous from the surface along the depth by the use of water-permeable forms [Ishihara et al. 1987] or the application of an inorganic impregnant to densify the microstructure of surface concrete. When the quality of surface concrete is altered from that of internal concrete in this manner, it is not clear whether or not the progress of carbonation can be appropriately expressed by putting the coefficient for concrete quality in Eq. (2) as  $\beta < 1$  or  $\beta > 1$ .

When applying finishing mortar or using cementitious stay-in-place forms [Baba et al. 2001], the carbonation of core concrete may progress after such mortar or forms have been carbonated, if they are monolithically integrated. Investigation has also been insufficient as to whether or not the progress of concrete carbonation can be appropriately expressed by putting the coefficient for the finishing material in Eq. (2) as  $\delta < 1$  or  $\delta > 1$ .

Meanwhile, Baba et al.[1987] proposed a method of estimating the carbonation depth of concrete having various surface layers including a finishing material, and several analyses have been conducted using the proposed equations [Kawanishi et al. 2006 & Kono et al. 2006]. The equations proposed by Baba et al. were derived by schematically solving the problem while introducing the concept of equivalent time.

In this study, the authors investigated the progress of concrete carbonation when the quality of surface concrete differs from that of internal concrete and when a cementitious material is monolithically integrated with base concrete. This was carried out theoretically using a model that expresses a phenomenon in which CO<sub>2</sub> steadily diffuses through the finishing material and concrete, obeying Fick's law, and instantaneously reacts with Ca(OH)<sub>2</sub>, which is present at the boundary between carbonated and noncarbonated regions. This causes the boundary to progressively shift deeper inward. This paper proposes a simple method of calculating such progress of carbonation. Note that the diffusion of CO<sub>2</sub> through mortar and concrete is assumed to be steady, though it is actually unsteady.

## 2 ANALYSIS

### 2.1 Subjects Of Analysis

The following two cases are investigated as subjects of analysis:

Case 1: Where finishing mortar is applied to concrete surfaces. A cementitious material in contact with concrete surfaces (e.g., stay-in-place forms) can be similarly dealt with.

Case 2: Where the microstructure near the surfaces is altered from that in internal concrete. This includes denser surfaces due to modification and rougher surfaces due to deterioration.

These two are basically similar, because in both cases a cementitious material having qualities different from internal concrete forms the surface layer, which is carbonated prior to the carbonation of internal concrete. The difference is that in Case 2 the depth of the surface cementitious layer is included in the cover depth whereas in Case 1 it is not.

Note that the case where a polymeric finishing material or coating is applied to concrete surfaces should be investigated separately, as carbonation of such finishing is irrelevant. Such a case is therefore not dealt with in this paper.

### 2.2 Case 1 (where finishing mortar is applied to concrete surfaces)

#### 2.2.1 Mortar Carbonation Phase

Figure 1 shows a pattern diagram expressing the diffusion of CO<sub>2</sub> and its reaction with Ca(OH)<sub>2</sub> in mortar in the phase of carbonation within finishing mortar. Assuming the coordinate of a concrete surface and the thickness of finishing mortar to be 0 and *d*, respectively, the coordinate of the surface of finishing mortar is  $-d$ . Putting the coordinate of the carbonation depth of finishing mortar as *x*, the amount of CO<sub>2</sub> reaching a depth of *x* during time  $\Delta t$  through a plane with an area of *S* perpendicular to the depth direction from the surface of finishing mortar,  $\Delta CO_2$ , is expressed by Eq. (3). If CO<sub>2</sub> reaching the depth of *x* instantaneously reacts with Ca(OH)<sub>2</sub>, then the amount of CO<sub>2</sub> consumed is expressed by Eq. (4).

$$\Delta CO_2 = D_m \cdot \frac{C_0}{x - (-d)} \cdot S \cdot \Delta t \quad (3)$$

$$\Delta CO_2 = H_m \cdot S \cdot \Delta x \quad (4)$$

where *x* : carbonation depth of finishing mortar ( $-d \leq x \leq 0$ , mm)

*t* : time(years)

*C*<sub>0</sub> : CO<sub>2</sub> concentration at the surface of finishing mortar

*D*<sub>*m*</sub> : diffusion coefficient of carbonated region in finishing mortar (mm<sup>2</sup>/year)

*S* : surface area(mm<sup>2</sup>)

*H*<sub>*m*</sub> : Ca(OH)<sub>2</sub> content per unit volume of finishing mortar

$\Delta x$  : thickness of boundary region

$\Delta t$  : time increment

By rearranging Eqs. (3) and (4),  $(x+d) \frac{\Delta x}{\Delta t} = \frac{D_m \cdot C_0}{H_m}$

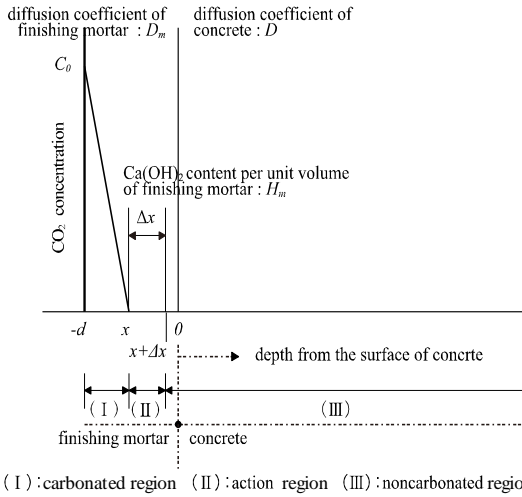
Assuming that  $\Delta t$  approaches zero, the following differential equation is obtained:

$$(x+d) \frac{dx}{dt} = \frac{D_m \cdot C_0}{H_m}$$

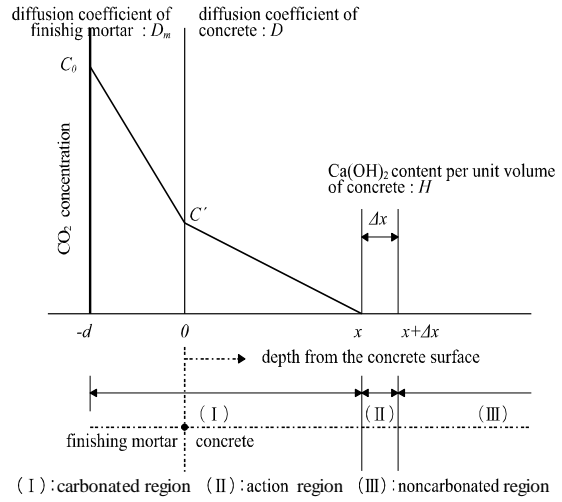
Both members are integrated as follows:  $\frac{1}{2} x^2 + d \cdot x = \frac{D_m \cdot C_0}{H_m} t + I_1$  (*I*<sub>1</sub>: integration constant)

When  $t=0$ ,  $x=-d$ . Therefore,  $I_1 = -\frac{1}{2} d^2$

$$\frac{1}{2} x^2 + d \cdot x - \frac{D_m \cdot C_0 \cdot t}{H_m} + \frac{1}{2} d^2 = 0$$



**Figure 1.** Model of CO<sub>2</sub> diffusion and reaction with Ca(OH)<sub>2</sub> while carbonation of finishing mortar is in progress.



**Figure 2.** Model of CO<sub>2</sub> diffusion while carbonation of concrete is in progress.

By solving this, 
$$x = \sqrt{\frac{2D_m \cdot C_0 \cdot t}{H_m}} - d \quad (5)$$

By putting  $\sqrt{\frac{2D_m \cdot C_0}{H_m}} = A'$ , Eq. (5) can be summarized as follows:

$$x = A'\sqrt{t} - d \quad (6)$$

Also, when finishing mortar is entirely carbonated, i.e., when  $x=0$ ,  $t = \frac{H_m}{2D_m \cdot C_0} d^2$

### 2.2.2 Concrete Carbonation Phase

Figure 2 shows a pattern diagram expressing the diffusion of CO<sub>2</sub> while concrete carbonation is in progress after the finishing mortar has been entirely carbonated. Assuming that carbonation has progressed to a depth of  $x$  from the concrete surface at an age of  $t$ , Eq. (7) expresses  $\Delta CO_2$ , the amount of CO<sub>2</sub> that diffuses through the finishing mortar layer and reaches the concrete surface during time  $\Delta t$  through a plane with an area of  $S$ , which is perpendicular to the depth direction. Similarly, Eq. (8) expresses  $\Delta CO_2$ , the amount of CO<sub>2</sub> reaching a depth of  $x$  from the concrete surface during time  $\Delta t$ . CO<sub>2</sub> diffusing to the depth of  $x$  is consumed by reaction with Ca(OH)<sub>2</sub>. The amount of CO<sub>2</sub> consumed by this reaction,  $\Delta CO_2$ , is expressed by Eq. (9), and then the CO<sub>2</sub> concentration becomes zero.

$$\Delta CO_2 = D_m \cdot \frac{C_0 - C'}{d} \cdot S \cdot \Delta t \quad (7)$$

$$\Delta CO_2 = D \cdot \frac{C'}{x} \cdot S \cdot \Delta t \quad (8)$$

$$\Delta CO_2 = H \cdot S \cdot \Delta x \quad (9)$$

where  $x$  : depth of concrete carbonation ( $0 \leq x$ , mm)

$C'$  : CO<sub>2</sub> concentration at the boundary between finishing mortar and concrete

$D$  : diffusion coefficient of CO<sub>2</sub> in carbonated concrete (mm<sup>2</sup>/year)

$H$  : Ca(OH)<sub>2</sub> content per unit volume of concrete

From Eqs. (7) and (8), 
$$C' = \frac{D_m \cdot x}{D_m \cdot x + D \cdot d} C_0 \quad (10)$$

From Eqs. (8) and (9), 
$$\frac{\Delta x}{\Delta t} = \frac{D \cdot C'}{H \cdot x} \quad (11)$$

By deleting  $C'$  from Eqs. (10) and (11) and assuming that  $\Delta t$  approaches zero, the following differential equation is obtained:

$$\left(x + \frac{D}{D_m} d\right) \frac{dx}{dt} = \frac{D \cdot C_0}{H}$$

By integrating both members,

$$\frac{1}{2} x^2 + \frac{D \cdot d}{D_m} x = \frac{D \cdot C_0}{H} t + I_2 \quad (I_2: \text{integration constant}) \quad (12)$$

Because  $t = \frac{H_m}{2D_m \cdot C_0} d^2$  when  $x=0$ ,  $I_2 = -\frac{D \cdot H_m \cdot d^2}{2D_m \cdot H}$

$$\text{By substituting } I_2 \text{ into Eq. (12), } \frac{1}{2} x^2 + \frac{D \cdot d}{D_m} x - \frac{D \cdot C_0}{H} t + \frac{D \cdot H_m \cdot d^2}{2D_m \cdot H} = 0 \quad (13)$$

$$\text{By solving Eq. (13), } x = \sqrt{\frac{2D \cdot C_0}{H} \left( \sqrt{\left(t - \frac{H_m \cdot d^2}{2D_m \cdot C_0}\right) + \frac{H \cdot D \cdot d^2}{2D_m^2 \cdot C_0}} - \sqrt{\frac{H \cdot D \cdot d^2}{2D_m^2 \cdot C_0}} \right)} \quad (14)$$

By putting  $\sqrt{\frac{2D \cdot C_0}{H}} = A$ ,  $\sqrt{\frac{H \cdot D \cdot d^2}{2D_m^2 \cdot C_0}} = R$ , and  $\frac{H_m \cdot d^2}{2D_m \cdot C_0} = T$ , Eq. (14) can be summarized as

$$\text{follows: } x = A \left( \sqrt{(t-T)+R^2} - R \right) \quad (15)$$

$$\text{Also, } A \cdot R = \frac{D \cdot d}{D_m}$$

Equation (15) is the same as the equation proposed by Baba et al. when  $T=R^2$ , i.e., when  $H_m \cdot D_m = H \cdot D$ .

### 2.3 Case 2 (where the microstructure of surface concrete is altered from that of internal concrete)

#### 2.3.1 Surface concrete carbonation phase

Figure 3 shows a pattern diagram expressing the diffusion of  $\text{CO}_2$  and its reaction with  $\text{Ca(OH)}_2$  while carbonation proceeds in surface concrete. The coordinate of concrete surface and the thickness of surface concrete are assumed to be zero and  $d$ , respectively. The amount of  $\text{CO}_2$  that diffuses through concrete and reacts with  $\text{Ca(OH)}_2$  at the boundary between carbonated and noncarbonated regions,  $\Delta \text{CO}_2$ , is expressed similarly to Case 1.

$$\Delta \text{CO}_2 = D_s \cdot \frac{C_0}{x} \cdot S \cdot \Delta t \quad (16)$$

$$\Delta \text{CO}_2 = H_s \cdot S \cdot \Delta x \quad (17)$$

where  $x$  : carbonation depth of surface concrete ( $x \leq d$ , mm)

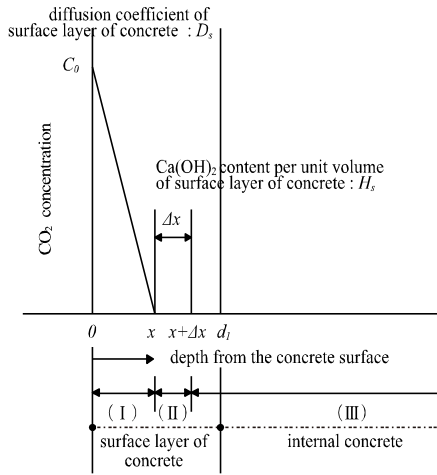
$D_s$  : diffusion coefficient for the carbonated region in surface concrete ( $\text{mm}^2/\text{year}$ )

$H_s$  :  $\text{Ca(OH)}_2$  content per unit volume of surface concrete

$$\text{This is solved as } x = \sqrt{\frac{2D_s \cdot C_0 \cdot t}{H_s}} \quad (18)$$

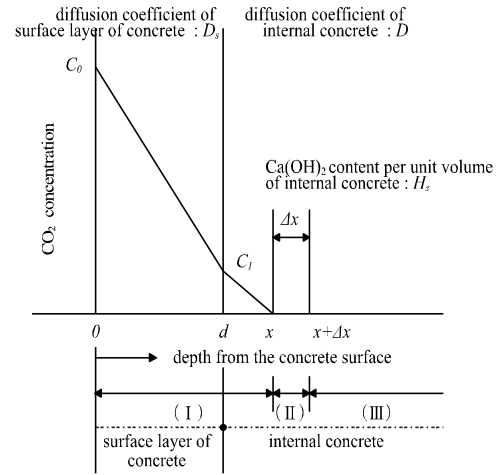
By putting  $\sqrt{\frac{2D_s \cdot C_0}{H_s}} = A''$ , Eq. (18) can be summarized as follows:  $x = A'' \sqrt{t}$

Also, when surface concrete is entirely carbonated, i.e., when  $x=d$ ,  $t = \frac{H_s}{2D_s \cdot C_0} d^2$



( I ) : carbonated region ( II ) : action region ( III ) : noncarbonated region

**Figure 3.** Model of CO<sub>2</sub> diffusion and reaction with Ca(OH)<sub>2</sub> while carbonation of surface concrete is in progress.



( I ) : carbonated region ( II ) : action region ( III ) : noncarbonated region

**Figure 4.** Model of CO<sub>2</sub> diffusion while carbonation of internal concrete is in progress.

### 2.3.2 Internal concrete carbonation phase

Figure 4 shows a pattern diagram expressing the diffusion of CO<sub>2</sub> in the phase in which carbonation proceeds in internal concrete after surface concrete has been entirely carbonated. The amount of CO<sub>2</sub> that diffuses through surface and internal concretes and reacts with Ca(OH)<sub>2</sub> at the boundary between carbonated and noncarbonated regions,  $\Delta CO_2$ , is expressed similarly to Case 1.

$$\Delta CO_2 = D_s \cdot \frac{C_0 - C'}{d} \cdot S \cdot \Delta t \quad (19)$$

$$\Delta CO_2 = D \cdot \frac{C'}{x - d} \cdot S \cdot \Delta t \quad (20)$$

$$\Delta CO_2 = H \cdot S \cdot \Delta x \quad (21)$$

where  $x$  : carbonation depth of internal concrete ( $d \leq x$ , mm)

$C'$  : CO<sub>2</sub> concentration at the boundary between surface and internal concrete

$D$  : diffusion coefficient of CO<sub>2</sub> in the carbonated region of internal concrete (mm<sup>2</sup>/year)

$H$  : Ca(OH)<sub>2</sub> content per unit volume of internal concrete

$$\text{From Eqs. (19) and (20), } C' = \frac{D_s(x-d)}{D_s x + (D - D_s)d} C_0 \quad (22)$$

$$\text{From Eqs. (20) and (21), } \frac{\Delta x}{\Delta t} = \frac{D \cdot C'}{H(x-d)} \quad (23)$$

By deleting  $C'$  and assuming that  $\Delta t$  approaches zero in Eqs. (22) and (23), the following differential equation is obtained:

$$\left\{ x + \frac{(D - D_s)d}{D_s} \right\} \frac{dx}{dt} = \frac{D \cdot C_0}{H}$$

$$\text{By integrating both members, } \frac{1}{2} x^2 + \frac{(D - D_s)d}{D_s} x = \frac{D \cdot C_0}{H} t + I_3 \quad (I_3: \text{integration constant}) \quad (24)$$

$$\text{Because } t = \frac{H_s}{2D_s \cdot C_0} d^2 \text{ when } x=d, \quad I_3 = \frac{(2D \cdot H - D \cdot H_s - D_s \cdot H)d^2}{2D_s \cdot H}$$

$$\text{By substituting } I_3 \text{ into Eq. (24), } x^2 + \frac{2(D - D_s)d}{D_s} x - \frac{2D \cdot C_0}{H} t - \frac{(2D \cdot H - D \cdot H_s - D_s \cdot H)d^2}{D_s \cdot H} = 0 \quad (25)$$

$$\text{By solving Eq. (25), } x = \sqrt{\frac{2D \cdot C_0}{H}} \left\{ \sqrt{\left( t - \frac{H_s \cdot d^2}{2D_s \cdot C_0} \right) + \frac{H \cdot D \cdot d^2}{2D_s^2 \cdot C_0}} - \sqrt{\frac{H \cdot D \cdot d^2}{2D_s^2 \cdot C_0}} \right\} + d \quad (26)$$

By putting  $\sqrt{\frac{2D \cdot C_0}{H}} = A$ ,  $\sqrt{\frac{H \cdot D \cdot d^2}{2D_s^2 \cdot C_0}} = R$  and  $\frac{H_s \cdot d^2}{2D_s \cdot C_0} = T$ , Eq. (26) can be summarized as follows:

$$x = A \left( \sqrt{(t-T) + R^2} - R \right) + d \quad (27)$$

Also,  $A \cdot R = \frac{D \cdot d}{D_s}$

Equation (27) is the same as the equation proposed by Baba et al. when  $T=R^2$ , i.e., when  $H_s \cdot D_s = H \cdot D$ .

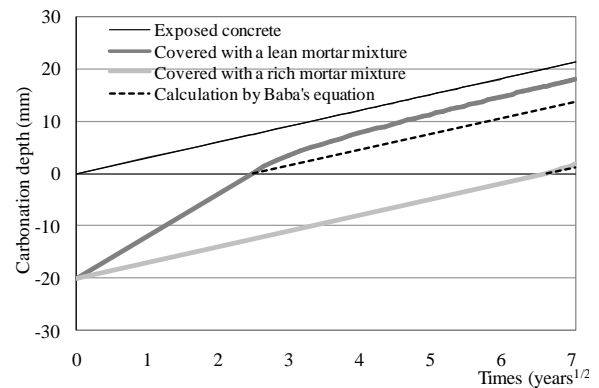
### 3 EXAMPLE OF NUMERICAL CALCULATION

In this section, a simulation of carbonation progress is attempted by substituting specific values into the above-mentioned analysis results where finishing mortar is applied to concrete surfaces. Table 1 gives the calculation conditions. Figures 5 and 6 show the progress of carbonation in concretes finished with mortars with a W/C of 50% and 60%, respectively.

**Table 1. Calculation conditions**

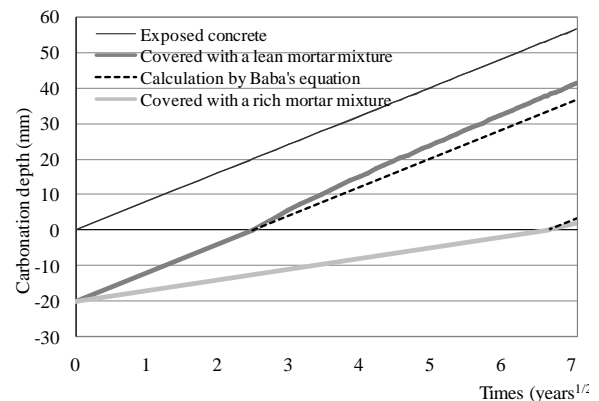
Concrete		Mortar coating		T	R
W/C	A	d	A'		
50	3	20	8	6.25	0.63
		20	3	44.44	4.44
65	8	20	8	6.25	1.67
		20	3	44.44	11.85

With a W/C of 50% (Fig. 5), carbonation of exposed concrete reaches a depth of around 21 mm after 50 years. When covered with a rich mortar mixture, carbonation reaches the bottom of the mortar layer after 44 years. With a W/C of 65% (Fig. 6), the carbonation depth in exposed concrete is 30 and 40 mm after 14 and 25 years, respectively. When covered with a lean mortar mixture, the carbonation depth in concrete is 30 and 40 mm after 33 and 48 years, respectively. When a rich mortar mixture is applied, carbonation remains within the mortar layer up until 44 years.



**Figure 5. Carbonation progress (W/C 50%).**

When comparing the calculations by Baba's equation and the present equation proposed by the authors, their differences are small regarding concrete covered with a rich mortar mixture, because of the slow progress of carbonation. With a lean mortar mixture, however, both equations lead to different results, as carbonation progresses in proportion to the square root of time by Baba's equation, whereas the present equation incorporates the effect of CO<sub>2</sub> permeation through carbonated mortar. With a W/C of 50%, for instance, it takes 35 years for carbonation to reach a depth of 10 mm by Baba's equation, whereas the present equation calculates this as 22 years. Thus the differences between both equations are found to be large in regard to a lean mortar mixture.



**Figure 6. Carbonation progress (W/C 65%).**

## 4 CONCLUSION

The progress of carbonation in concrete in which the quality of the surface region differs from that of internal concrete was theoretically investigated based on pseudo steady diffusion. A method of calculating such progress was proposed, and carbonation was simulated. The results are summarized as follows:

- (1) After carbonation of the surface layer is completed, carbonation in internal concrete proceeds according to the equations below respectively when finishing mortar or a cementitious material is applied to the concrete surface (Eq.15) and when the microstructure of surface concrete differs from that of internal concrete (Eq.27).
- (2) The equations proposed by the authors led to results slightly different from those proposed by Baba et al., as the present equations incorporate the effect of CO<sub>2</sub> permeation through carbonated mortar or surface concrete. These are valid when assessing the effect of the quality of finishing materials and surface concrete.

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