Investigation in Mathematical Models of Chloride Diffusion Coefficient in Concrete Exposed to Marine Environment

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Abstract: Degradation of RC (reinforced concrete) in maritime structures has become a worldwide problem due to its excessive costs of maintenance, repair and replacement in addition to its environmental impacts and safety issues. Degradation of both concrete and steel which is the main reason of reduction in the service life of RC structures strongly depends on the diffusion process of moisture and aggressive species. In this paper, the major and popular mathematical models of diffusion process in concrete are surveyed and investigated. Predominantly in these models, the coefficient of chloride diffusion into the concrete is assumed to be constant. Whereas, experimental records indicate that diffusion coefficient is a function of time. Subsequently, data analysis and comparisons between the existing analytical models for predicting the diffusion coefficient with the existing experimental database are carried out in this study. Clearly, these comparisons reveal that there are gaps between the existing mathematical models and previously recorded experimental results. Perhaps, these gaps may be interpreted as influence of the other affecting parameters on the diffusion coefficient such as temperature, aggregate size and relative humidity in addition to the water cement ratio. Accordingly, the existing mathematical models are not adequate enough to predict the diffusion coefficient precisely and further studies need to be performed.

Key words: Chloride diffusion coefficient, corrosion of steel in concrete, mathematical model of diffusion.

1. Introduction

Reinforced concrete has been used for construction of infrastructures around the globe due to its durability, good compressive strength and relatively lower cost. However, diffusion of aggressive ions such as chloride into the concrete can cause severe deterioration of structure due to corrosion of embedded steel reinforcing. Corrosion of reinforcing steel in concrete caused by chloride transport in concrete in marine environment has received increasing attention in recent years because of its widespread occurrence and high cost of maintenance and repair [1, 2]. Reinforced concrete structures are generally exposed to various environmental

conditions during their lifetime. Therefore, degradation of materials and subsequently premature deterioration may occur which can significantly affect the service life of the structures [3].

According to the vast investigations, it is found out that the dominant factor of degradation process is the chloride-induced corrosion of the steel reinforcement in concrete [4, 5]

According to the recent intensive study commissioned by the United States FHWA (Federal Highway Administration), the annual direct cost of corrosion in the United States was 276 billion dollar in 1998, or 3.1% of the GDP (gross domestic product) [6]. Cost analyses of corrosion were also conducted in other countries, such as the United Kingdom, Japan, Australia and Kuwait. Even though the level of effort varies greatly among these studies, all of them have estimated the total annual cost of corrosion as ranging

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between 1% and 5% of each country's GNP (gross national product).

2. Mathematical Models of Diffusion

There are a few number of mechanisms such as diffusion, absorption, migration in an electrical field by which chloride transport can take place in concrete.

Diffusivity is the relationship between the concentration flux J of diffusing material and the gradient of the chemical potential that is assumed to drive the mass diffusion process. Most of the conducted studies on diffusion-based mathematical models are supported by Fick's law of diffusion. Fick's law relates the diffusive flux to the concentration field, by postulating that the flux goes from the regions of high concentration to the region of low concentration, with a magnitude that is proportional to the concentration gradient (spatial derivative) dimension.

Fick's second law can be expressed as follows:

$$\frac{\partial \phi}{\partial t} = D \frac{\partial^2 \phi}{\partial x^2} \quad (1)$$

where, ϕ is the concentration rate of diffusive flux (mol/m^3), t is the time (s), D is the diffusion coefficient (m^2/s) and x is the location (m).

If we assume that the diffusion coefficient is not constant and depends upon the coordinate and/or concentration ratio, Fick's second law can be defined as Eq. (2):

$$\frac{\partial \phi}{\partial t} = \nabla (D \cdot \nabla \phi) \quad (2)$$

Since the chloride ions diffusion into RC (reinforced concrete) structures is the main cause of deterioration of RC structures, the present study will focus on this phenomenon. Generally, due to chloride diffusion, a concentration gradient develops near the concrete surface. Thus, the time, at which the critical chloride content (threshold value) reaches the steel surfaces and depassivates it, can be regarded as the initiation time of corrosion. The gradient of chloride

content is often described by an error function model (RILEM, 1996) which fulfils the condition of Fick's second law of diffusion as shown in Eq. (3):

$$C_{(x,t)} = C_s \left[1 - \text{erf} \left[\frac{x}{2(Dt)^{\frac{1}{2}}} \right] \right] \quad (3)$$

where $C_{(x,t)}$ is the chloride content (gradient) at depth x at time t ($\text{\%/m}^3$), C_s is the chloride concentration at the concrete surface ($\text{\%/m}^3$), x is the depth from concrete surface (m), D is the diffusion coefficient (m^2/s), erf is the error function and t is time (s).

The Fick's law is used to describe the diffusion process based on two assumptions firstly, the fluxing species must not react with the matrix and secondly the matrix is homogenous in structure and composition.

Recently, an inclusive ion transport process for predicting of the chloride ingress into reinforced concrete structures has been described by the Nernst-Planck equation [7]. This model, which is well established based on the traditional electrochemical theorem [8], describes the ion transport as a combination of three different mechanisms including: diffusion, convection (carried along by moisture movement, i.e., by absorption or hydraulic pressure), and electrical migration.

Kropp [9] stated that depending on the environmental exposure conditions as well as on the moisture content of the concrete element, the ingress of chlorides into concrete occurs through coupled multiple transport mechanisms. However, most models for chloride ingress in concrete are still based on the Fick's second law of diffusion as the main transport mechanism. Moreover, diffusion-based models such as Refs. [10, 11] have been found broadly acceptable for long-term monitoring of chloride ingress in concrete structures.

Basheer et al. [12] expressed their mathematical model based on Fick's law and incorporated the quantity of chloride binding with the matrix to the equation. Applying the principle of mass conservation

to the concentration of chloride ion couple utilizing Fick's diffusion law yields:

$$\frac{\partial C}{\partial t} = \text{div} [D_i \nabla (C)] - \frac{1}{\omega} * \frac{\partial S}{\partial t} \quad (4)$$

where, C is the free chloride concentration in the porous media (mol/cm^3), S is the quantity of bound chloride ions (mol/cm^3), ω is the content of water in which diffusion occurs (per unit mass of cement), and D_i is the chloride diffusion coefficient in the pore solution (cm^2/s).

Since the diffusion and concentration of chloride ions into concrete and on the reinforcement surface are long-term process, using the proposed potential for accelerated diffusion will be established to determine the diffusivity at any time. Liang et al. [13], by using equation of Tang and Nilsson [14] and considering the influence of the added potential for acceleration, suggested the following expression based on Fick's second law:

$$J_{ix} = -D_e \left[\frac{\partial C_f}{\partial x} - \frac{eFE}{RT} C_f \right] \quad (5)$$

where, J_{ix} is the flux of chloride ion, D_e is the effective diffusion coefficient for the free chloride ion, C_f is the concentration of free chloride ion (mol/L), e is the valency of ion, F is the Faraday's constant ($96,500 \text{ C/mol}$), E is the electrical field density (V/m), $R = 8.314 \text{ (J/mol}\cdot\text{K)}$ the ideal gas constant, and T is the absolute temperature (K).

The total chloride ion concentration C_t in $\delta V = A \times \delta x$ can be obtained from Eq. (6) as follows:

$$C_t = C_b + \varepsilon \cdot d \cdot C_f \quad (6)$$

where, C_b is the bound chloride ion (mol/L), ε the total porosity of concrete, and d the degree of saturation by the pore water solution.

The equation of continuity for the chloride ion is presented by Eq. (7) and shown in Fig. 1:

$$\delta(C_t) A \cdot \delta x = J_x \cdot A \cdot \delta x - J_{(x-\delta x)} \cdot A \cdot \delta x \quad (7)$$

By expanding the Taylor's series and substituting in Eq. (7), it can be rewritten as:

$$\frac{\partial (C_b + \varepsilon C_f)}{\partial t} = \frac{\partial}{\partial x} \left[D_e \left(\frac{\partial C_f}{\partial x} - \frac{eFE}{RT} C_f \right) \right] \quad (8)$$

Fig. 1 Scheme for the equation of the continuity of chloride ion.

Assuming that C_f and C_b are parts of a reversible process, the forward and backward absorption rates can be written as Eq. (9) and Eq. (10), respectively:

$$r_a = k_a \cdot \varepsilon \cdot d \cdot C_f \quad (9)$$

$$r_b = k_b \cdot C_b \quad (10)$$

where, k_a and k_b are forward and backward deposition coefficient, respectively. The net absorption rate is shown in Eq. (11):

$$\frac{\partial C_b}{\partial t} = r = r_a - r_b = k_a \cdot \varepsilon \cdot d \cdot C_f - k_b \cdot C_b \quad (11)$$

Eqs. (10) and (11) yield:

$$\frac{\partial (\varepsilon d C_f)}{\partial t} = \frac{\partial}{\partial x} \left[D_e \left(\frac{\partial C_f}{\partial x} - \frac{eFE}{RT} C_f \right) \right] - r \quad (12)$$

If both porosity and the effective diffusion coefficient for mature concrete are independent of space and time, that is, both ε and D_e are constant with respect to x and t , then Eq. (12) can be rewritten as:

$$\varepsilon \frac{\partial C_f}{\partial t} = D_e \left[\frac{\partial^2 C_f}{\partial x^2} - \frac{eFE}{RT} \frac{\partial C_f}{\partial x} \right] - r \quad (13)$$

where, $C = \frac{C}{C_{en}}$, $\omega = \frac{C_b}{C_{bsat}}$, $z = \frac{x}{L}$,

$$\tau = \frac{D_e}{\varepsilon d L^2} \cdot t, \quad \lambda = \frac{eFE L}{RT}, \quad \beta = \frac{C_{bsat}}{\varepsilon d C_{en}}.$$

C_{en} is the chloride concentration at the interface of the concrete surface to the external environment (surface chloride concentration), C_{bsat} is the maximum concentration that can be absorbed by the concrete, and L is the cover thickness of concrete.

From Eqs. (11)-(13), the non-dimensional Eqs. (14) and (15) can be driven:

$$\frac{\partial C}{\partial \tau} = \frac{\partial^2 C}{\partial z^2} - \lambda \frac{\partial C}{\partial z} - \beta \frac{\partial \omega}{\partial \tau} \quad (14)$$

$$\beta \frac{\partial \omega}{\partial \tau} = \alpha C - \mu \omega \quad (15)$$

where, $\alpha = \frac{k_a L^2 \varepsilon}{D_e}$ and $\mu = \frac{k_b L^2 C_{bsat}}{D_e C_{en}}$.

Eqs. (14) and (15), which were driven simultaneously from the diffusion of accelerated chloride ions in an electric field and the absorption in concrete pores, α , β , λ and μ are all constant with respect to z and τ .

If $C_b \rightarrow 0$ then $\omega \rightarrow 0$, Eq. (15) should be modified as follows:

$$\beta \frac{\partial \omega}{\partial \tau} = \alpha C \quad (16)$$

Substituting Eq. (16) into Eq. (14), the governing Eq. (17) can be obtained:

$$\frac{\partial C}{\partial \tau} = \frac{\partial^2 C}{\partial z^2} - \lambda \frac{\partial C}{\partial z} - \alpha C \quad (17)$$

Finally, Liang et al. [13] expressed the exact analytical solution by using Laplace transform equation as following:

$$C_{(z,\tau)} = \frac{1}{2} \exp \left\{ \left[\frac{\lambda}{2} - \sqrt{\frac{\lambda^2}{4} + \alpha} \right] z \right\} \cdot \left\{ \begin{aligned} & \operatorname{erfc} \left[\frac{z}{2\sqrt{\tau}} - \sqrt{\left(\alpha + \frac{\lambda^2}{4} \right) \tau} \right] + \\ & \exp \left(z \sqrt{4\alpha + \lambda^2} \right) \cdot \\ & \operatorname{erfc} \left[\frac{z}{2\sqrt{\tau}} + \sqrt{\left(\alpha + \frac{\lambda^2}{4} \right) \tau} \right] \end{aligned} \right\} \quad (18)$$

$\operatorname{erfc}(x)$ is called the complementary error function and is defined as Eq. (19):

$$\operatorname{erfc}(x) = 1 - \operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_x^\infty e^{-u^2} du \quad (19)$$

Many researchers have used Fick's second law to predict the initiation time of chloride induced corrosion with a given diffusion coefficient D and

surface chloride content C_s , based on the assumption that cement matrix in concrete is chemically inert and evenly porous [15, 16]. However, these hypothesis and models have been challenged since [10] addressed the time-dependent characteristic of chloride transport in terms of D and C_s . Later, Bentz et al. [17] study confirmed the change of D with time.

Crank [18] proposed their model based on the relation between the diffusion and electro-migration of ions in the same electrolyte. Based on Nernst-Einstein equation, if concrete is considered to be a solid electrolyte, the diffusion of charged species i in concrete is related to its partial conductivity σ_i .

$$D_i = \frac{RT\sigma_i}{Z_i^2 F^2 C_i^2} \quad (20)$$

where, D_i is the diffusivity of species i (cm^2/s), R the ideal gas constant ($8.314 \text{ J/mol}\cdot\text{K}$), T the absolute temperature (K), σ_i the partial conductivity of species i (S/cm), Z_i the charge of species i , F is Faraday's constant ($96,500 \text{ C/mol}$), C_i is the concentration of species i (mol/cm^3).

If the partial conductivity σ_i and the concentration C_i are determined, the diffusivity of species i , D_i can be calculated from Eq. (20).

However, the partial conductivity σ_i can be defined as:

$$\sigma_i = \sigma \times t_i \quad (21)$$

where, σ is the conductivity of concrete, and t_i is the transference number of species i , which can be defined as:

$$t_i = \frac{Q_i}{Q} = \frac{I_i}{I} \quad (22)$$

where, Q_i and I_i are the electric quantity and current contribution of species i respectively, to the total electric quantity Q and current I .

If the diffusivity of an ion is to be determined first the Nernst-Einstein equation, and second the transference number of this ion should be known. A simple and proper approach is to make the transference number approximately equal to 1.0. For example, if the concrete is filled with a concentrated salt solution, the transference number may be assumed

as 1.0, and C_i may be considered equal to the chloride concentration in the pore solution, or in the fill the concrete. When $T = 298$ K and $C_{cl^-} = 0.05 \text{ mol/cm}^3$. In this model also the chloride diffusion coefficient has been assumed as a constant factor.

3. Mathematical Model of Chloride Diffusion Coefficient

The main weakness and imperfection of the transport process models is the assumption of constant diffusion coefficient for concrete. However, experimental data illustrates that the diffusion coefficient is a function of time.

Mangat and Molloy [19] have proposed the following mathematical relationship to determine the diffusion coefficient as a time-dependent parameter. In this model, the water cement ratio is the only assumed influencing factor on the diffusion coefficient.

$$D_t = D_i \times t^{\left[0.6-25\left(\frac{w}{c}\right)\right]} \quad (23)$$

where, D_i is the chloride diffusion coefficient in the pore solution.

Song et al. [20] introduced their mathematical model based on the time-dependent diffusion coefficient and surface chloride concentration phenomenon shown by Eq. (12). In fact, capillary pores reduce when they are partially blocked as the cement matrix hydrates [21], which implies loss of paths for chloride ions.

$$D(t) = D_0 \left(\frac{t_0}{t} \right)^m \quad (24)$$

where, D_t is the diffusion coefficient at time t (m^2/s), D_0 the diffusion coefficient at time t_0 (m^2/s), t_0 the standard time (one year or 28 days), t is time (s), and m is a constant.

4. Database Analysis and Discussion

To conduct analysis, the proposed mathematical model (Eq. (23)) by Mangat and Molloy [19] was selected to compare to available experimental

database in literature (Table 1). Based on database, the analysis was performed for various water cement ratio including 0.32, 0.4, 0.54, 0.58 and 0.7.

For every water cement ratio, the magnitude of the chloride diffusion coefficient in different times determined by Eq. (23) and compared to the database experimental results.

Calculated theoretical values and experimental results of chloride diffusion coefficient versus time are presented in Figs. 2-6.

In Figs. 2 and 3, for the concrete with W/C ratios 0.32 and 0.4, respectively, there exist a gap approximately 1×10^{-12} (m^2/s) between the theoretical model and tests results.

For water cement ratio of 0.4 the trend is same but the gap has been increased and mathematical model shows lower diffusion coefficient compare to experimental data. If a service life is design based on the mathematical model, it may show a longer service life than the actual condition of the structure.

In addition, comparison of the diagrams shown in Figs. 3-5 reveals that by increasing the W/C ratio in

Table 1 Database for diffusion coefficient.

Diffusion coefficient vs. time			
W/C	$T(\text{day})$	$D (*10^{-12})$	Reference
0.32	1	7.39	Luping and Nilsson (1992)
0.32	3	6.22	
0.32	7	4.93	
0.32	28	3.22	
0.32	90	1.79	
0.32	180	1.74	
0.4	7	3	Polder (1995)
0.4	28	2	
0.4	98	1	
0.54	7	4	Luping and Nilsson (1992)
0.54	28	3	
0.54	98	2.5	
0.58	28	52.3	Mangat and Molloy (1994)
0.58	90	23.8	
0.58	270	10	
0.7	1	45.6	Luping and Nilsson (1992)
0.7	3	26.95	
0.7	7	21.1	
0.7	28	14.5	
0.7	90	15.3

Fig. 2 Chloride diffusion coefficient vs. time for concrete with W/C of 0.32.

Fig. 3 Chloride diffusion coefficient vs. time for concrete with W/C of 0.4.

Fig. 4 Chloride diffusion coefficient vs. time for concrete with W/C of 0.54.

excess of 0.4, the existing gap between mathematical model and experimental results increases. For instance, when we increase W/C ratio from 0.4 to 0.7 the existing gap increases from 1×10^{-12} to 15×10^{-12} for 90 days (Fig. 5) and from 1×10^{-12} to 30×10^{-12} for 28 days (Fig. 4).

Fig. 5 Chloride diffusion coefficient vs. time for concrete with W/C of 0.58.

Fig. 6 Chloride diffusion coefficient vs. time for concrete with W/C of 0.7.

This study clearly illustrates that up to $W/C = 0.5$, the differences between the mathematical model and experiment results are negligible while the mathematical model shows higher diffusion coefficient. However, beyond $W/C = 0.5$, the differences are getting increased by incising the W/C ratio while the experimental results exhibit higher diffusion coefficient.

Also, a review on the experimental results indicate that in some cases with reducing the W/C ratio, diffusion coefficient increases, e.g., compare the graphs $W/C = 0.32$ and $W/C = 0.4$, which theoretically, it has to be reduced. This phenomenon proves that there are other influencing factors on diffusion coefficient which has to be taken into account such as proportion and maximum size of coarse aggregate. In order to obtain an accurate mathematical model these factors should be considered as well.

5. Summary and Conclusions

The major source of degradation and deterioration of RC structures is due to the diffusion of chloride into concrete. Many comprehensive studies and investigations have been already conducted to model mathematically this phenomenon. Most of these models are based on Fick's law which assumes that diffusion coefficient is constant and independent of time. Whereas, the experimental results are shown that the diffusion coefficient is a time-dependent parameter.

These mathematical models have been summarized in Table 2.

Two proposed mathematical models by Mangat (Eq. (23)) and Song (Eq. (24)) of diffusion coefficient as a time-dependent parameter were analyzed and compared with experimental database. The presented comparative study on the transfer process of the chloride diffusion versus time, exhibits the gaps between analytical and experimental results (Figs. 2-6).

Table 2 Summary of the mathematical models for chloride diffusion into concrete.

Mathematical model	Presented by reference
$\frac{\partial \phi}{\partial t} = D \frac{\partial^2 \phi}{\partial x^2}$	Fick's second law D, constant
$\frac{\partial \phi}{\partial t} = \nabla \cdot (D \cdot \nabla \phi)$	Fick's second law D, depend on the concentration
$C_{(x,t)} = C_s \left[1 - \operatorname{erf} \left[\frac{x}{2(Dt)^{\frac{1}{2}}} \right] \right]$	RILEM Report 14 D, constant
$J_{ix} = -D_e \left[\frac{\partial C_f}{\partial x} - \frac{eFE}{RT} C_f \right]$	Liang et al. [13] Added potential
$D_i = \frac{RT\sigma_i}{Z_i^2 F^2 C_i^2}$	Crank [18] D, depends on partial conductivity of species
$\frac{\partial C}{\partial t} = \operatorname{div}[D_i \nabla(C)] - \frac{1}{\omega} * \frac{\partial S}{\partial t}$	Basheer et al. [12] Effect of chloride binding capacity (free chloride)
$D_t = D_i \times t^{\left[0.6-25\left(\frac{w}{c}\right)\right]}$	Mangat and Molloy [19] D, a function of W/C ratio
$D_{(t)} = D_0 \left(\frac{t_0}{t} \right)^m$	Song et al. [20]

These gaps reveal that influence of the other responsible parameters in addition to water cement ratio such as aggregate size, temperature and humidity must be included precisely in the mathematical models.

To predict the service life of RC structures accurately, the deviation of the chloride diffusion coefficient with time should be determined. Subsequently, an appropriate mathematical model has to be proposed to model the diffusion coefficient at any given time intervals accurately.

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