Review on Chloride Ingress in Concrete: Chloride Diffusion and Predicting Corrosion Initiation Time

Pradeep K. Goyal[1], Andualem E. Yadeta[2]

[1,2]*Department of Civil Engineering, Delhi Technological University, Delhi, India*

Abstract - Corrosion is a common cause of degradation of reinforced concrete (RC) structures. Even though various reasons cause the phenomenon, chloride-induced corrosion in concrete is an important issue in current research. The maintenance of corroded RC increases the cost of facilities; durability assessment is helpful for economic savings and the performance of structures. Therefore, this paper reviews the chloride diffusion process and the time it takes for corrosion to start in RC structures. In this paper, Fick's second law is considered for the transport mechanism of chlorides into concrete structures. Different models for predicting corrosion initiation time by previous scholars are critically reviewed.

Keywords - Concrete structures, Durability, Chloride diffusion, Reinforcement bar, Corrosion.

1. Introduction

Concrete durability is vital for the economy and the safety of society. However, the performance of constructed facilities can be affected by strength degradation over time. Among the common degradation mechanisms in concrete structures, rebar corrosion is a significant issue. Corrosion may occur by transporting different agents like chloride ions into concrete. Chlorides can present in the fresh concrete mix when chloride-contaminated materials are used during concrete mixing, and they can also diffuse into the hardened concrete at a later stage. When RC structures are exposed to seawater or de-icing salts such as calcium chloride ($CaCl_2$) and sodium chloride ($NaCl$), steel passivity will be broken, risk steel reinforcement cross-section loss. Corrosion risk increases as chloride concentration rise; however, it can be mitigated by using supplemental cementitious elements in concrete (Zhou et al., 2014). It is also further stated that including silica fume and corrosion inhibitors in the concrete mix helps speed up the onset of corrosion in concrete (Dinh 2017). Even though structures are designed for a specific service period, corrosion in concrete degrades the durability of concrete structures, and they fail to provide the required service. To protect against rebar corrosion in concrete, identifying the transport mechanism of chlorides is helpful (Liu, Easterbrook, and Li, 2017).

Chlorides can penetrate the concrete by the diffusion transport mechanism, assuming that the concrete cover is in a fully saturated environmental condition. This general diffusion process in materials is described using Fick's second law (Que 2007). Among the previous works done to predict corrosion initiation time in concrete reported by different authors, most models were developed based on concrete microstructure (Jaturapitakkul, Cheewaket, and Chalee 2014). The diffusion process in concrete structures can be affected by different environmental factors, and there is a need to review the available models for the corrosion process in RC structures.

Therefore, the objective of this study is to review the process of chloride diffusion and determine the appropriate model to predict the corrosion initiation time in concrete. First, the chloride diffusion mechanisms are investigated using Fick's second law. Secondly, the most suitable model for predicting corrosion initiation time in concrete was determined after a thorough assessment of several models proposed by previous scholars.

1.1. Steel Corrosion

Corrosion is a gradual disintegration of materials (mostly metals) caused by chemical and/or electrochemical reactions with their environment, resulting in mass loss and dimension changes (ASTM G193-12d 2012). Corrosion requires water, oxygen, and ions which are all present in the environment (Palanisamy 2019). The general steel corrosion process can be briefly explained using a simple model (see Fig. 1).
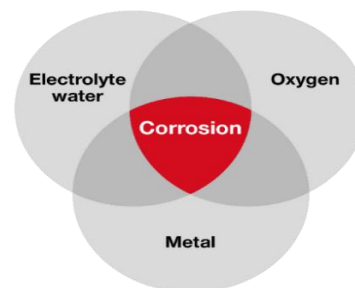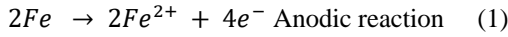

Fig. 1 Corrosion process

The reinforcing steels used in concrete structures have a passive coating on their surface that protects them from corrosion. Because the pH of concrete is very high, around 13, in mild ambient circumstances and the passive protective

layer inhibits corrosion initiation. The change of steel into rust is an electrochemical process in nature, commonly called electrochemical reaction since it is different from the chemical reaction as it involves the passage of electrical charges.

Corrosion is a two-stage electrochemical process in general: primary and secondary process. In the primary process, the steel dissolves in the pore water solution:

$$2Fe \rightarrow 2Fe^{2+} + 4e^- \text{ Anodic reaction} \quad (1)$$

To keep the electrical neutrality of the reaction process, the electrons released in the primary process should be utilized, and then the secondary electrochemical process occurs on metals:

$$4H^+ + 4e^- \rightarrow H_2 \text{Cathodic reaction} \quad (2)$$

Therefore, corrosion starts in the secondary stage of the electrochemical process. Many different products can be formed based on influencing factors like the pH of the solution and the environmental condition. The secondary reaction process consumes the Fe^{2+} produced in the primary process. Then it initiates corrosion by taking oxygen and hydrogen from pore water, as illustrated in Fig. 2below:
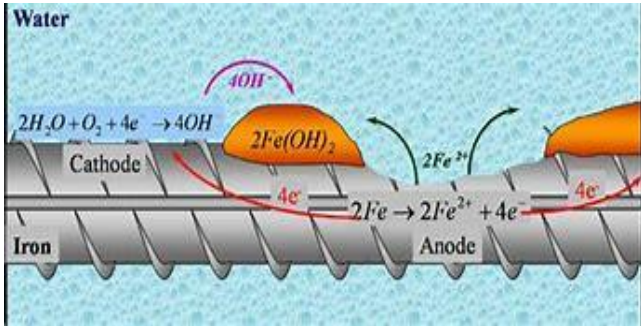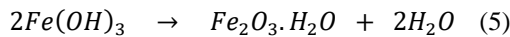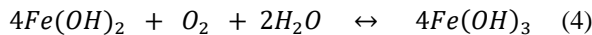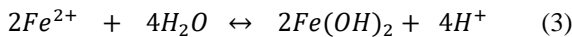

Fig. 2 Corrosion cell (Portland Cement Association 2019)

$$2Fe^{2+} + 4H_2O \leftrightarrow 2Fe(OH)_2 + 4H^+ \quad (3)$$

$$4Fe(OH)_2 + O_2 + 2H_2O \leftrightarrow 4Fe(OH)_3 \quad (4)$$

$$2Fe(OH)_3 \rightarrow Fe_2O_3.H_2O + 2H_2O \quad (5)$$

The corrosion product $Fe(OH)_2$ will be further oxidized to Fe_2O_4 or Fe_2O_3 in the usual circumstances of steel embedded in concrete. The equations in 1-5 are universal equations that can be applied to any metal corrosion process. In this study, chloride-induced steel corrosion is a specific scenario in which chlorides penetrate concrete from the environment and cause corrosion by lowering the pH and breaking the passive layer of the bar.

1.2. Corrosion Influencing Factors in Concrete
Corrosion can be influenced by different factors that determine structures' performance against a corrosive

environment. These can be categorized as practices during design, construction, and environmental factors. The construction practice determines concrete's durability properties (permeability, porosity, etc.). For example, appropriate curing decreases the porosity, which determines the durability of concrete against aggressive environmental actions (Koleva et al. 2008). The design practice significantly influences the durability of concrete in a corrosive environment (Alonso and Sanchez 2009). The penetration of chlorides into concrete can be minimized with a lower w/c ratio and increased concrete strength (Liu, Hu et al., 2020). According to (Kristawan et al. 2017), the larger concrete cover increases the time for rebar in structures to start rusting. The performance of concrete structures against corrosion also depends on environmental conditions such as temperature and humidity.

2. Chloride-Induced Concrete
Even though there are many aggressive agents in the environment, the exposure of RC structures to chlorides is the principal cause of reinforcement corrosion. Chlorides can be dissolved in water and diffuse through concrete or reach the steel via pore structures and micro-cracks (Tang and Gan 2015). Chloride-containing additives can also cause corrosion. The chloride ions contained in de-icing salts and seawater can cause corrosion when oxygen and moisture are present, as illustrated in Fig. 3below. Chlorides in concrete can also be facilitated by cement, water, aggregate, and admixtures.

Chloride ions develop an oxide film over the reinforcing steel, which speeds up corrosion reactions and reduces structural durability (Valipour et al., 2013). Even though the chlorides penetration process is complex, diffusion is the most common transport mechanism used to assess the durability of concrete (Liu, Easterbrook, and Li, 2017).

Fig. 3 An RC structure exposed to chlorides (Legault 2011)

The mechanism by which chlorides cause corrosion is unknown. Still, the most widely accepted explanation is that chlorides penetrate the protective layer of the reinforcement

more easily than other ions, exposing the rebar to corrosion. Then the corrosion products continue to expand, causing component cracking and failure (Chen, Baji, and Li, 2018). It is shown in Fig. 4 below that chloride-induced corrosion is occurred due to exposure of an RC structure to seawater.

Fig. 4 Chlorides-induced corrosion (Graham 2017)

The chloride-induced reinforcement corrosion in RC structures is essentially an electrochemical reaction process where the passive protective layer of steel is lost due to chloride ions forming microcells on the reinforcement surface. The electrochemical process is initiated when moisture in the pores of concrete functions as an electrolyte and the area next to the chloride ion concentration acts as a cathode (Zhang, Zhang, and Ye, 2018).

3. Chloride Diffusion

Diffusion is the movement of any substance from a region of a high concentration area to a low concentration area along a concentration gradient(Pal, Paulson, and Rousseau 2013). It is a very important phenomenon as many reactions are diffusion-dependent processes. In the case of diffusion of chlorides in concrete, only the free chlorides contribute to the reaction potential. In this process, free chlorides initiate corrosion while the bound chlorides can be released (Luque et al., 2014). The concentration gradient and chlorides diffusion result from a high concentration region of chlorides. As a result, chloride diffusion is determined by absorption for a particular depth into concrete, and diffusion becomes the major transport mechanism in concrete (Hunkeler 2005). The typical chloride diffusion process in concrete is illustrated in Fig. 5 below.

The diffusion process generally occurs under the steady-state and the non-steady-state conditions. Under the steady-state condition, the chloride concentration in the diffusion process is constant, whereas it is time-dependent under the non-steady-state condition. Under the steady-state conditions, diffusion responds to the concentration gradient, which is expressed as the changes in concentration due to the changes in position, $\partial c/\partial x$. This diffusion process is commonly expressed using Fick's first law as follows:
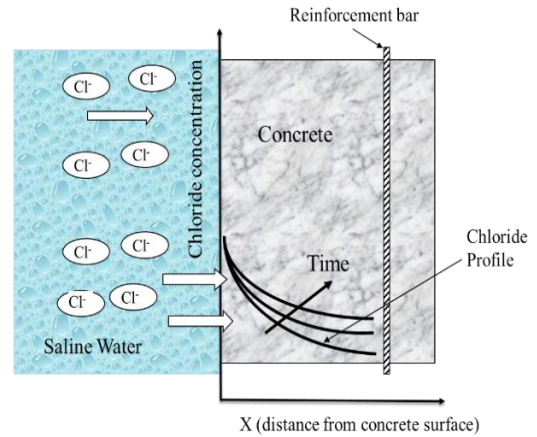

Fig. 5 Typical chloride ions diffusion process in RC

$$J = -D_e \frac{\partial c}{\partial x} \qquad (6)$$

Where:
J is the diffusion flux (flow rate of ions) of the system (cm^2/s);
D_e is the coefficient of diffusion or diffusivity for Fick's 1st law (cm^2/s), and
$\partial c/\partial x$ is the gradient concentration. If linear $\partial c/\partial x = \Delta C/\Delta X = C_2 - C_1/X_2 - X_1$.

Flow occurs at the negative concentration gradient, as shown by the negative signal in the equation. In Fick's first law, the diffusion coefficient, D_e is called the 'effective' diffusion coefficient. This coefficient measures concrete penetration in relation to the flow rate of chlorides in concrete. (Tang, Nilsson and Basheer 2011). Under the steady conditions, diffusion is calculated using Fick's second law. Fick's second law is found in the first law to determine how scattering causes a change in the focus of an object in relation to time. Since concrete is an equally permanent method in the filling area, Fick's second law is often used to describe the process of chloride diffusion in concrete (Martin-Perez et al., 2000, Yuan et al., 2009, Wang, Gong, and Wu, 2019):

$$\frac{\partial c}{\partial t} = -D_a \frac{\partial^2 c}{\partial x^2} \qquad (7)$$

where:
c is the concentration of chlorides (cm^3)
t is the exposure duration (s);
x is the depth (cm); and
D_a is the diffusivity for Fick's second law, also called the 'apparent diffusivity' (cm^2/s).

Several attempts were made to determine the apparent diffusion coefficient in Equation 7 to determine the corrosion initiation time (Garboczi and Bentz 1992,

Ababneh, Benboudjema and Xi 2003, Oh and Jang 2004, Bentz 2007, Han 2007, Wang and Ueda 2011, Li, Xia and Lin 2012, Zhang, Dong and Jiang 2013). Most of them reported that the apparent difference could be obtained by inserting the error line function of Fick's second law into the chloride profile of the particular concrete being investigated. (Wang, Gong, and Wu 2019). The apparent diffusivity cannot be considered a parameter, but it is a regression coefficient that measures the permeability of a particular concrete exposed to a chloride-rich environment(Tang, Nilsson, and Basheer 2011). The difference between the apparent diffusion and the effective diffusion coefficient is that the apparent diffusivity includes the binding capacity of concrete in Fick's second law, but the first law does not. Chloride binding plays a crucial role in the chloride diffusion process in concrete. The relationship between the two coefficients can be determined from Fick's second law as given in equation (8) below:

$$D_a = \frac{D_e}{P_{sol}\left(1+\frac{\partial C_b}{\partial c}\right)} \qquad (8)$$

Where p_{sol} is the porosity of the concrete.

4. Prediction of Corrosion Initiation Time in Concrete

Mathematically, the time to start corrosion in concrete can be calculated based on the diffusion parameters, especially can be taken from Fick's second law. This is because of the difference in the concentration of the chlorides from the structure's surface penetration through the concrete into the reinforcement surface. If the rebar area's chloride ions exceed the concrete's critical threshold and destroy the resulting layer, Fick's second rule applies, and then corrosion begins. The corrosion rate of the reinforcement is heavily employed in RC structural evaluations, maintenance decisions, and residual life forecasts. (Yu et al. 2014). Therefore, most of the mathematical equations developed for durability assessment of RC structures are related to steel corrosion following a simple model that was developed by (Tuutti 1980)in the very back period in which corrosion mechanism is divided into two phases, as shown in Fig. 6below.
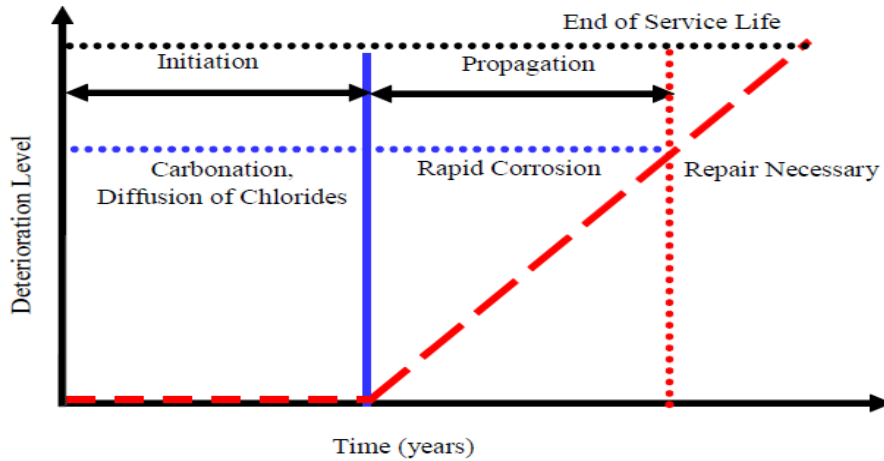

Fig. 6 Tutti's model (Andrade 2020)

$$t = t_i + t_p \qquad (9)$$

Where t_i is the initiation period for corrosion
t_p is the propagation period for corrosion

The initiation time is the amount of time it takes for chlorides on the surface of the concrete to permeate into the micropores. When the concentration of chlorides reaches a threshold level, steel reinforcement de-passivates, and the corrosion start phase terminates. The propagation period then begins, during which a chemical reaction occurs that causes the reinforcing steel to rust.

4.1. Fick's Second Law of Diffusion
The passage of chloride ions into structures is the primary cause of rebar corrosion in concrete. As a result,

based on Equation 7 of Fick's second law, the following formula can be used to calculate the rebar corrosion initiation time in concrete (Bazant 1979):

$$t_{ic} = \frac{x_c^2}{4D_c}\left[erf^{-1}\left(1-\frac{C_{cr}}{C_o}\right)\right]^{-2} \qquad (10)$$

From previous studies, the mathematical equations to determine chlorides diffusion into concrete as a depth and time function are computed using Fick's second law (Equation 7), as discussed in the previous section. From this equation, the corrosion initiation time can be calculated. The solution to this one-dimensional diffusion problem of a specific material depends on boundary conditions and certain assumptions.

4.2. Constant Chloride Diffusion

Chloride diffusion is a complex process as the transport of chlorides is specifically affected by other ions present in concrete pore solution. The following general solution was proposed by most studies, assuming constant chloride diffusivity and chloride concentration at the surface of RC structures. Fick's second law leads to the following equation when these parameters are treated as constants (Anacta 2009):

$$C(x,t) = C_i + (C_{sa} - C_i)erfc\left[\frac{x}{\sqrt{4(t - t_{ex})D_a}}\right] \quad (11)$$

where:

$C(x,t)$ is the chloride concentration at time t and depth x
C_i is the initial chloride concentration (constant depth).
C_{sa} is the chloride concentration on concrete surface
D_a is the apparent coefficient of diffusion (constant)
x is the depth from the surface
t is the inspection time

We can solve (Equation 11) by taking D as a constant over time, and the surface chloride concentration is constant; we have (Crank 2004):

$$C(x,t) = C_s\left[1 - erf\left(\frac{x}{2\sqrt{Dt}}\right)\right] \quad (12)$$

where:

$C_{x,t}$ is the concentration of chlorides at depth x and time t,
C_s is the concentration of chlorides on concrete surfaces,
C_o is the initial uniform concentration of chlorides at the surface,
x is the depth from the surface, t is time, and
D is the apparent coefficient of chloride diffusion.

When it comes to concrete, applying the constant chloride diffusivity concept is limited to a particular structure with long exposure to environmental actions. This is because, at significantly longer exposure of structures to chlorides, the chloride diffusivity of the concrete exhibits a constant character.

4.3. Time-dependent chloride diffusion

The chloride concentration varies with time because the diffusion rate determines the amount of diffusing chloride ions into concrete at a given depth. As the exposure duration increases, the chlorides ion concentration in concrete drops. According to (Poulsen 1993), an equation for the determination of a time-dependent diffusion coefficient (D_a) is given by:

$$D_a = \frac{1}{t}\int_0^t D(t)dt \quad (13)$$

Different scholars previously developed many models. Among the different models, the model created by (Anacta 2009) for corrosion initiation time prediction considers some environmental factors such as temperature, humidity, and rainfall in his assumption. This model also further considered the influence of the duration of concrete exposure to environmental actions. In this model, the equation to calculate the depth of chloride diffusion, which helps to compute corrosion initiation time, was formulated as shown in (Equation 14)below:

$$x_c = 2s\sqrt{D_c t} \quad (14)$$

The chloride diffusion coefficient (D_c), which is taken as the function of environmental factors, is given by:

$$D_c = D_{c,rmt}\, x f_1(t) x f_2(T) x f_3(RH) x f_4(R) \quad (15)$$

In this expression, $D_{c,rmt}$ is called the reference coefficient of chloride diffusion, which can be obtained from the laboratory test. To critically review the previous models reported by different authors to predict corrosion initiation time are summarized (see Table1).

The chloride diffusion and the associated process in concrete depend on the environmental condition that the structure is exposed to (Bester 2014). The previous (Equation 14) is more suitable for concrete structures when the depth of chloride diffusion is equivalent to the depth of concrete cover to predict when chloride ions reach the rebar surface. The calculated time, t, as shown in the following (Equation 16), also helps to determine the shape factor, S, and the diffusion coefficient, D_c, which are significant parameters in the computation of corrosion initiation time in concrete.

$$t_{ic} = \frac{f_s}{D_c}\left[\frac{X_c}{2S}\right]^2 \quad (16)$$

$$f_s = 0.316\xi\sqrt{t_{ic,ref}} \quad (17)$$

where:

t_{ic} is the corrosion initiation time (years)
$t_{ic,\,the\,ref}$ is the reference corrosion initiation time which will be obtained from experiments (days)
x_c is the thickness of concrete cover (mm)
f_s is the reinforcement factor
ξ is the curve-fitting parameter

From previous works of different authors, Equation 16 for the determination of corrosion initiation time in concrete is found as the best applicable equation due to the fact that the performance of RC structures depends on environmental conditions. The big advantage of using this formula is the application of local data for validation, which considers the environmental factors, including temperature, humidity, and rainfall, as these are the significant factors for corrosion.

Table 1. Comparison of different models for prediction of corrosion initiation time in concrete

Model	Basis	Equation
Clear's Model (Clear 1976)	Empirical	$t_{ic} = \dfrac{129.\, x_c^{1.22}}{(\frac{w}{c}).\,[C_s]^{0.42}}$
Bazant's Model (Bazant 1979)	Fick's second law	$t_{ic} = \dfrac{x_c^2}{4D_c}[erf^{-1}\,(1 - \dfrac{C_{cr}}{C_o})]^{-2}$
Poulsen-Mejlbro's Model (Poulsen 1993)	Fick's second law	$t_{ic} = t_{ex}\, X \left(\dfrac{0.5 x_c}{\sqrt{t_{ex}D_{aex}}}\right)^{\frac{2}{1-a}} X \left((\dfrac{1}{inv\,\Lambda p\,(y_{cr)}})^{\frac{2}{1-a^-}}\right)$
Yamamoto-Hosoya's Model (Yamamoto and Hosoya 1995)	Fick's second law	$t_{ic} = \dfrac{1}{D_c}[\dfrac{x_c}{2erf^{-1}\,(1 - C_{cr}/C_o)}]^2$
Anacta's Model (Anacta 2009)	Fick's second law	$t_{ic} = \dfrac{f_s}{D_c}\left[\dfrac{x_c}{2S}\right]^2$
Tang-Nilsson's Model (Tang and Nilsson 1992)	Numerical	$Q_{i,j}(total) = Q_{i,j-1}(total) + Q_{i,j}(diff)$

5. Conclusion

The required parameters for predicting chloride diffusion and the corrosion initiation time in concrete structures are all given in the critical review and can be determined experimentally. The chloride binding capacity inside the concrete and the chloride diffusion coefficient decreases over time. This implies that the equation that considers the influence of chloride binding and the chloride diffusion coefficient forecasts more accurate performance for structures than Fick's second law equation. Compared to other models, the equation for corrosion initiation time computation that considers the local environmental conditions and materials (Equation 16) is more appropriate. Therefore, the model formulated in Equation 16 is recommended to determine the corrosion initiation time of reinforcement bars with very high accuracy in concrete structures.

6. Declaration of Competing Interest

The authors declare that they have no known personal or financial conflicts of interest that could influence their work.

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