CHLORIDE DIFFUSION IN CONCRETE AND ITS IMPACT ON CORROSION OF REINFORCEMENT

Abul K. Azad

Department of Civil Engineering
King Fahd University of Petroleum and Minerals
Dhahran 31261, Saudi Arabia

ABSTRACT

Chloride-induced corrosion of reinforcement is a major problem for concrete durability in a salt-laden environment. For a concrete with negligible amount of initial chloride inherited at the construction stage, the gradual buildup of the required amount of chloride to initiate corrosion of reinforcement takes place predominantly through diffusion of chloride ions from external sources under a concentration gradient. Consequently, diffusion of chloride ions in concrete has received a great deal of interest. Most of the researchers have modeled the chloride ingress by Fick's law of diffusion, advocating its general applicability to concrete.

In this paper, the parameters influencing chloride diffusion and the general applicability of Fick's law are reviewed, highlighting the impact of chloride diffusion coefficient on corrosion. Laboratory generated test data on concrete samples made with local aggregates shows that chloride diffusion coefficient for local concrete appears to be high and it is adversely impacted by the thermal effects. The results also show that Fick's law can be used to model diffusion and to predict corrosion initiation time with reasonable degree of accuracy.

KEYWORDS

Diffusion; corrosion; chlorides; cement content; durability.

INTRODUCTION

The single-most dominant factor of deterioration of reinforced concrete structures in the coastal region of the Gulf States is irrefutably identified as the chloride-induced corrosion of reinforcement. The concomitant presence of hot, humid climatic conditions and high level of airbourne and waterbourne chlorides have in effect created an aggressive

environment for concrete construction, which is highly conducive to corrosion of reinforcement (Rasheeduzzafar et al., 1984).

Chloride-induced corrosion of reinforcement can take place in the presence of oxygen and moisture when the chloride buildup within the structures exceeds a threshold value. Even for a carefully constructed concrete with negligible or practically no chloride inherited at the construction stage, the gradual buildup of the required level of chloride content to initiate corrosion of reinforcement takes place slowly through ingress of chloride ions from external sources.

The transport phenomenon associated with the movement of chloride ions in structures exposed to salt-laden environment is attributed mostly to diffusion of chloride ions into a porous concrete under a concentration gradient. Chloride diffusion coefficient of a concrete which depends upon the pore structure of the concrete, characterizes this flow under a given concentration of chloride exposure and is considered as a characteristic property of a hardened concrete.

In this paper, the parameters affecting the chloride diffusion are reviewed by briefly presenting the salient features of the past voluminous work in this area and following this, its importance is highlighted in estimating the corrosion initiation time under a given exposure condition. Fick's second law of diffusion (Crank, 1975) has been widely regarded as an acceptable basis to model the chloride ingress in concrete. The results of recently conducted tests on concrete samples using local aggregates (Navaz, 1994; Sharif et al., 1997b) have further endorsed the viewpoint that Fick's law of diffusion can be used in ideal conditions as the predictive model for a fair estimation of corrosion initiation time and have clearly demonstrated that chloride diffusion coefficient is adversely influenced by the hot weather conditions.

CHLORIDE DIFFUSION IN CONCRETE

General

It appears that Collepardi et al. (1972) first calculated meaningfully the diffusion coefficient from laboratory tests for various cement paste mixes using Fick's second law of diffusion (Eq. 1) and concluded that chloride penetration proceeds by ionic diffusion. Gjorv and Vennesland (1979) studied the diffusion of chloride ions into concrete from seawater. The test variables were w/c ratio, type of cement, cement content, polarization potential and aggregate type. Results showed that porosity and permeability, which increase with w/c ratio, affect the diffusion only in the exterior layers while in the interior, diffusion was affected by chloride binding and ion exchange. The lower diffusion in blended cements was attributed to the lesser amount of calcium hydroxide which means a lesser capacity for ion exchange and therefore lesser penetration of chlorides. The tricalcium aluminate (C_3A) was seen to have no significant effect on chloride diffusion if its percentage was less than 8.6%.

The study of diffusion through hardened cement pastes of various compositions received considerable attention (Page et al., 1981; Goto and Roy, 1981; Page et al., 1986; Diab et al., 1988). Increase in diffusion rate with increased w/c is noted. Page et al. (1986) concluded that sulphate resisting cement performs poorly against corrosion and diffusion of chlorides.

The effect of silica fume and fly ash on diffusion characteristics was explored by Byfors (1987). Midgley and Illston (1984) investigated the effect of chloride concentration by considering two solutions of NaCl with different concentrations. Test results showed that the depth of penetration at a given time increases with w/c ratio and the concentration of chloride ions. The study also concluded that the presence of chloride ions alters the pore-size distribution of the hardened cement paste and smaller pores are associated with higher chloride ions. Ost and Monfore (1986) looked into the chloride migration rate into aggregates of concrete by comparing the rate of migration in concrete and cement paste. Experimental data showed that chloride migrated into concrete more readily than that into the cement paste.

To study the effects of different types of salts, Hansson et al. (1985) used calcium chloride, sodium chloride and potassium chloride as exposures. Results showed that, of the three salts, calcium chloride had the most deleterious effect. It was also inferred that the total chloride ion content of the pore solution does not alone account for the corrosion rate, but the rate of corrosion is also controlled by other factors that include porosity, pH level and the availability of oxygen.

Applicability

Concerns have been raised with regard to the applicability of diffusion as an appropriate modeling of chloride ingress, as there are several mechanisms by which chloride ions can be transported within the concrete (Arup, 1983; West and Hime, 1985). Several processes may be combinedly operative, making it difficult to identify a single mechanism. One of the mechanisms for surface penetration is intrusion of chloride-bearing water into capillary pores of unsaturated (dry) concrete by capillary action. Alternate wetting and drying can lead to buildup of chloride ions through absorption. If a structure is not dried to a high degree for a prolonged period of time, chloride penetration into concrete by absorption and capillary suction is basically restricted to a small depth below the surface. If there is a differential head of chloride-bearing water, permeability will also influence the ingress of chlorides for which higher permeability coefficient will permit higher rate of flow. The other dominant mechanism of chloride transport is the diffusion which takes place under a concentration gradient. If the outside chloride concentration is higher than the inside of concrete, the migration of chloride ions through pore water in concrete will take place by diffusion.

The relative importance of the two major mechanisms of chloride transport, namely diffusion and absorption, depend on the moisture content of concrete. Absorption may be dominant if a dry concrete with significant loss of pore water is wetted with chloride-bearing water, whereas for a reasonably moist concrete (sufficient level of pore water exists) diffusion process will prevail under concentration gradient. However, researchers tend to agree that in most cases diffusion can be assumed to be the basic transport mechanism of chloride ions for reasonably moist structures (Browne and Geoghegan, 1985; Tuutti, 1985).

An argument against the validity of pore diffusion of chloride ingress is that some part of the chloride is chemically bound in concrete due to reaction with cement and remains as immobilized (Bruenfield, 1986; Verbeck, 1975). The chloride concentration profile therefore depends to some extent upon the type of cement used in concrete.

As part of the chloride in concrete is chemically bound due to reaction of chloride ions with constituents of cement, the free chloride concentration is of importance for corrosion initiation (Tuutti, 1985). Raharinaivo and Jean-Marie (1986) arrived at a formulation to find the apparent diffusivity considering the trapping or the reaction of the chloride ions with the constituents of cement. The study concluded, among others, that the chloride penetration can be modeled by Fick's diffusion law in most cases if other factors such as pressure, evaporation and frost action are neglected. However, when a cement has a high proportion of tricalcium aluminate content and when a concrete is of small porosity, Fick's law cannot be applied with full validity. Gau and Cornet (1985) have considered a reaction rate between chloride ions, cement paste and a chloride convection coefficient for the solution of Fick's law. However, as the convection effect over long term with large area is small, it can be ignored to obtain a reasonable solution of Fick's law (Funahasi, 1990). According to researchers (Gau and Cornet, 1985; Funahasi, 1990), the reaction between Cl^- and cement can be accounted for by considering an effective diffusion as $D_e = D/[(k+1)(k+p)]$, where k represents slope of chloride adsorption isotherm and p is the porosity of concrete.

From their study on chloride penetration and Cl^-/OH^- ratio in cement pastes, Kayyali and Haque (1988) concluded that the critical ratio of Cl^-/OH^- for depassivation exceeded the set limit of 0.6 reported by Raharinaivo and Jean-Marie (1986) and Hausmann (1967). The influence of w/c ratio, pozzolanic admixture and temperature on chloride ion diffusion was conducted by Weyers and Smith (1989) who used a non-linear regression analysis to determine diffusion coefficient. They concluded that Fick's law was applicable in all cases except for silica fume mixture. Relationship between w/c and diffusion coefficient was suggested for two exposure conditions.

The penetration of wind-blown chlorides into concrete was looked into by Jaegerman (1990) by considering the effect of w/c ratio and curing regimes on such penetration for specimens exposed to Mediterranean Sea conditions. This study showed that chloride penetration increases with an increase in w/c ratio and is influenced by the curing regime. Diffusion coefficients are calculated using Fick's law.

Dhir et al. (1991) from their tests on concrete cube specimens made out of ordinary Portland cement and cured under different conditions, found that the diffusivity calculated by applying Fick's law approached a value found by a rapid test devised by them. A method is suggested and nomograms are provided to estimate the time required to reach a certain chloride concentration for a constant diffusivity. In their study of diffusion of chloride ions and hydroxyl ions into concrete, Sergi et al. (1992) concluded that total and free chloride profiles could be interpolated to a good approximation by Fick's law. The study also confirmed that the relationship between free and bound chloride could be represented by Langmuir adsorption isotherm. Lately, Teng and Lee (1992) have proposed a different numerical technique to find apparent diffusion coefficient of chloride in concrete which is claimed to be more suitable for non-steady state of diffusion. The method uses nonlinear chi-square and Newton's (NCSN) method to determine the diffusion coefficient.

Solution of Fick's Second Law of Diffusion

Fick's second law of diffusion is given as

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} \qquad (1)$$

in which c = concentration, t = time, x = distance from the boundary and D is the diffusion coefficient. The solution of Eq. (1) for a *semi-infinite domain* with a *uniform concentration* of c_s at the surface ($x = 0$) is (Crank, 1975)

$$c_x = c_s\left[1 - \text{erf}\frac{x}{2\sqrt{D_e t}}\right] \qquad (2)$$

when c_x = concentration at distance x from the boundary, 'erf' is the error function and D_e is the effective diffusion coefficient. For a concrete member having an initial chloride content of c_i built-in at the construction stage, the diffusion Eq. 2 takes the form

$$\frac{c_x - c_i}{c_s - c_i} = 1 - \text{erf}\frac{x}{2\sqrt{D_e t}} \qquad (3)$$

For the application of Fick's second law of diffusion, several techniques have been used to find solution of the partial differential equation (Eq. 1). As the equation involves concentration of chloride ions c_s as ppm at the exposed surface (at $x = 0$) and the concentration at a distance x from the surface, c_x is conveniently measured as percentage by weight of cement (or concrete), the direct application of Fick's law is therefore not possible. Sergi et al. (1992) have measured chloride content as ppm by extracting the pore water in concrete. Weyers and Smith (1989) have adopted a nonlinear regression analysis and a finite difference method was used to determine diffusion coefficient by Funahasi (1990). Numerical integration is pursued by Midgley and Illston (1984) and an iterative program has been used to determine the value of diffusion coefficient which best fits the data of chloride concentration by Liam et al. (1992). Nagano and Naito (1985) provide a solution of Fick's law with stepwise uniform (periodic) functions for c_s at the boundary.

Measurement of Diffusion Coefficient

Generally, diffusion coefficients have been determined either by using the conventional method of subjecting specimens to known concentrations of chloride solutions and then chemically measuring the chloride concentration at different depths of the samples (e.g. Gjorv and Vennesland, 1979; Page et al., 1981; Dhir et al., 1991; Sergi et al., 1992) or by using diffusion cells. Each of these concentration gradient type tests requires a long duration, often upto 3-9 months to get meaningful results prompting researchers to look for rapid methods.

In order to expedite the diffusion process in a diffusion cell experiment, a small potential difference of 2 V (DC) has been applied as perturbance (Goto and Roy, 1981), on the assumption that such a small disturbance, which expedites diffusion, does not critically affect the measured values. Whiting's Coulomb test (Whiting, 1981) for rapid measurement of chloride permeability, developed for bridge deck testing, has been adopted by AASHTO Standard (1983). Unfortunately, this test does not provide information on diffusion of chloride ion itself in concrete.

Dhir et al. (1990) have proposed a simple rapid test using diffusion cell to determine diffusion coefficient. In this method, a small potential difference is applied at a low current across a concrete specimen in a diffusion cell. Even then, about a week would be required to obtain an estimate of diffusion coefficient from this experiment. Lately, Luping and Nilsson (1992) have proposed a new rapid method by applying an electric field by which chloride penetration profile can be obtained in much shorter time (several hours or a few days depending on the type of concrete). Several advantages of this method have been highlighted.

In a limited study on typical concretes of the Eastern Province of Saudi Arabia, the values of diffusion coefficients were determined using indoor and outdoor exposure conditions (Navaz, 1994; Sharif et al., 1997b). Results show that there is a dramatic increase in values of the diffusion coefficients and hence the rate of chloride ingress due to the outdoor exposure.

For the first time, Sharif et al. (1997a) have proposed a new method of determining chloride diffusion coefficient using the concept of bulk diffusion and Knudsen diffusion as applied to porous catalysts (Yang, 1987; Jackson, 1977). From the laboratory setup, the ratio of porosity to tortuosity of concrete which is considered as a constant for a porous solid medium, can be calculated and this then can be used to determine diffusion coefficient (Sharif et al., 1997a).

Field Studies

The interest of researchers to gather field data on the chloride penetration and to see how the penetration can be modeled has led to numerous studies of insitu chloride profiles for old concrete structures (West and Hime, 1985; Funahasi, 1990; Nagano and Naito, 1985; Liam et al., 1992; Masuda, 1987). From the best fitting plots of the measured data, diffusion coefficient of chloride was calculated. All have reaffirmed in principle the general applicability of Fick's law and based on insitu findings, advocated the use of diffusion law to predict corrosion initiation time for a given threshold value of chloride concentration (Funahasi, 1990; Liam et al., 1992). Nagano and Naito (1985) have considered three different types of chloride exposures at the boundary, including stepwise uniform concentration, and have provided the solution of Fick's law. Furthermore, they have examined the effect of coating on concrete on the chloride ingress. Masuda (1987) has considered the effect of chloride exposure on structures which are indirectly exposed to the seawater by taking into account the chloride concentration in atmosphere. Fick's law has been used again with new boundary condition.

USE OF CHLORIDE DIFFUSION COEFFICIENT

As the predominant mechanism of chloride transport in concrete is by diffusion, the effective chloride diffusion coefficient D_e of a concrete is viewed as a key indicator of its susceptibility to corrosion. The higher the value of D_e, the faster is the rate of diffusion of chloride ions and hence the chloride buildup in concrete.

The importance of chloride diffusion coefficient as a durability-based material property has received greater attention only after the revelation that chloride-induced corrosion is the major problem for concrete durability. More often in the past, permeability has been used as an indicator of the degree of vulnerability of a concrete to reinforcement

corrosion. Although both permeability and diffusion are related to flow, the fact that permeability is associated with flow through a porous medium under a pressure head and diffusion takes place under a concentration gradient, the latter is more appropriate to characterize the movement of chloride ions in concrete.

Fick's law of diffusion has been adopted widely as a reasonably accurate model for chloride ingress in concrete. However, the direct application of Eq. (3) for chloride diffusion is hindered by the intrinsic requirements of the applicable values of D_e and c_s. Although the value of D_e of a concrete can be measured with sufficient accuracy, the applicable value of c_s at the surface, which must be expressed as percentage weight of concrete, is difficult to ascertain.

For a practical application of Fick's equation, the difficulty is further compounded by the fact that the chloride exposure at the surface is rarely a constant and, more often, it fluctuates with seasonal and diurnal variation. For the direct application of Eq. (3), the commonly encountered case of non-uniform chloride concentration has to be replaced by an equivalent uniform exposure concentration of c_s or alternatively, the insitu chloride exposure can be replaced by an idealized periodic function of c_s and then Eq. (1) can be solved for the assumed boundary condition.

Prediction Model for Corrosion Initiation

Eq. (3) has been used widely as a predictive model for an estimation of corrosion initiation time for which the value of the required chloride concentration at the reinforcement level to initiate corrosion is needed. Despite a voluminous work on the required threshold value of chloride concentration for corrosion initiation, the unanimity on an universal value is far from close due to the complexity of the corrosion process, which depends upon a number of interactive factors including pH of concrete, cement content and the ratio of Cl^-/OH^- in pore solution (Hausmann, 1967; Comité Euro-International Du Beton, 1982; Rasheeduzzafar et al., 1984; Hansson and Sorensen, 1990; Clear and Hay, 1973; Stratfull et al., 1975; Browne, 1982; Hussain et al., 1996).

The reported values of the threshold level of chloride content vary from 0.6 kg/m^3 to 1.33 kg/m^3 of concrete. European state-of-the-art report concludes that there is no consensus on the permissible limits of chloride concentration but a concentration level of 0.35-1.0% by weight of cement may trigger corrosion (Comité Euro-International Du Beton, 1982). For the Middle East Environment, the reported range is 0.05-0.7% by weight of cement (Rasheeduzzafar et al., 1984). A review of suggested values indicates that, in general, corrosion may occur with chloride content exceeding 0.35% by weight of cement. ACI (1989) recommends a maximum limit of 0.3% by weight of cement for water-soluble chloride for reinforced concrete construction.

For a given structure, the depth of cover to steel reinforcement, d_c, is known. With the knowledge of the amount of chloride concentration, c_c, at the reinforcement level which is necessary to trigger corrosion, Eq. (3) leads to the following prediction model for estimation of corrosion initiation time t

$$\frac{c_c - c_i}{c_s - c_i} = 1 - \text{erf}\frac{d_c}{2\sqrt{D_e t}} \tag{4}$$

EXPERIMENTAL RESULTS USING LOCAL CONCRETE

With the aim of determining the applicable values of D_e for local concrete, an experimental program was undertaken using different mix designs with sulphate resisting cement (ASTM Type V). The reason behind the choice of this type of cement is that it is widely used in foundations and in near or below grade work and also it yields less chemical binding of chloride ions in view of its lower amount of C_3A, thus providing stronger validity to the application of Fick's law.

The values of D_e were calculated from experimental data obtained from two types of parallel tests: (a) traditional ponding with NaCl solution and (b) gas diffusion technique (Navaz, 1994; Sharif et al., 1997a). The details of experimental work are given by Navaz (1994). In order to examine the effect of hot climatic condition on D_e, outdoor exposure condition was used in parallel to the indoor exposure of controlled laboratory conditions. A typical plot of chloride concentration at 70 and 105 days of exposure in indoor environment is shown in Fig. 1.
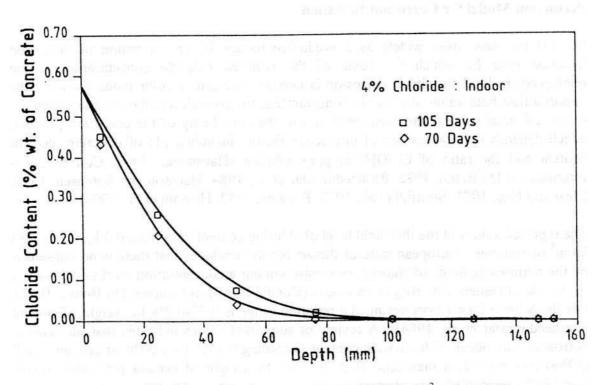

Fig. 1. Chloride profiles (cement content = 350 kg/m³; w/c = 0.7)

Some of the values of D_e are shown in Table 1 which lead to three important observations: (i) the values of D_e are generally higher for locally made concrete due to poor quality of aggregates, (ii) D_e increases rapidly with increasing water-cement ratio and (iii) most importantly, the outdoor environment (temperature ranging from 8 to 50°C)

sharply increases the value of D_e, a phenomenon attributable to the effect of temperature on kinetics of diffusion and associated factors (Navaz, 1994; Sharif et al., 1997b). The last finding casts an ominous sign for the use of laboratory generated D_e for members exposed to heat and cool cycles of outdoor environment.

Table 1. Effective diffusion coefficient D_e and surface concentration c_s
(cement content = 350 kg/m³)

W/C ratio	Cl concentration	Indoor		Outdoor	
		D_e (cm²/sec)	c_s (% wt. of conc.)	D_e (cm²/sec)	c_s (% wt. of conc.)
0.40	4%	8.03×10^{-8}	0.32	24.45×10^{-8}	0.36
	8%	8.52×10^{-8}	0.56	27.54×10^{-8}	0.75
0.55	8%	28.10×10^{-8}	0.76	73.50×10^{-8}	1.25
0.70	4%	68.45×10^{-8}	0.58	165.0×10^{-8}	0.83
	8%	66.30×10^{-8}	0.82	170.3×10^{-8}	1.95

The experimental results showed that D_e predominantly depends upon w/c ratios and cement content for identical concreting procedure. Consequently, the following expression of D_e was derived using a regression analysis of test data for indoor conditions.

$$D_e = \left[82.74 - 425.9\,(w/c) + 568.42\,(w/c)^2 + 4.26\,(\bar{c})^{-6} \right] 10^{-8} \qquad (5)$$

when $\bar{c}$ is the normalized cement content obtained by dividing the actual cement content used in kg/m³ by 350 kg/m³.

An effort was made to check the validity of Eq. (4) for the prediction of the corrosion initiation time for which an experimental program was undertaken using reinforced concrete blocks with known cover (Navaz, 1994). The corrosion activity was monitored using measurement of corrosion current density and half-cell potential measurements. The time at which corrosion of reinforcement was first noted was recorded as the 'corrosion initiation time'. Table 2 shows the values of laboratory observed corrosion initiation time and the predicted values from Eq. (4) using a value of $c_c = 0.35$ percent weight of concrete. A reasonable correlation between the actual and the predicted time can be observed from Table 2, considering the fact that the value of c_c which has a considerable influence on t is not exactly known.

CONCLUSIONS

Of all possible mechanisms of chloride ingress in concrete, diffusion is recognized as a predominant transport mechanism of chloride ions in moist concrete structures under a

concentration gradient. This has led to the advocation of Fick's law of diffusion as an applicable model for diffusion of chloride ions in concrete. The general adoptability of diffusion for concrete and the various factors which influence diffusion are reviewed, highlighting the recent awareness of the chloride diffusion coefficient as a characteristic parameter of concrete for chloride-induced corrosion of reinforcement.

Recent experimental investigation on local concrete has shown that chloride diffusion coefficient of local concrete is relatively higher and it increases sharply in hot weather conditions, a fact which should be reckoned with. Test data also shows that the prediction model for corrosion initiation time, based on Fick's law, can be used with reasonable accuracy.

Table 2. Comparison of actual and predicted time for corrosion initiation

W/C ratio	Chloride concentration in NaCl solution	Cover to bar (mm)	Value of D_e (cm²/sec × 10⁻⁸)	Value of c_s % wt of concrete	Corrosion Initiation Time in days	
					Predicted	Actual
0.55	4%	38	24.70	0.46	143	200
0.55	8%	38	24.70	0.76	100	120
0.55	8%	76	24.70	0.76	399	285
0.40	8%	38	7.59	0.56	380	280

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