# CHLORIDE DIFFUSION COEFFICIENT OF CONCRETE IN THE ARABIAN GULF ENVIRONMENT

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الخلاصة :

يُقيم هذا البحث عامل انتشار أيون الكلوريد في الخرسانة المصنوعة من الركام الجيري والإسمنت البورتلاندي المقاوم للسُلفات التي تستعمل بكثرة في منطقة الخليج العربي. تم دراسة المتغيرات التالية: (١) نسبة الماء للإسمنت، (٢) كمية الإسمنت، (٣) تركيز نسبة الكلوريد على سطح الخرسانة، (٤) الأجواء المحيطة. ومن ثَمَّ مُحاولة إيجاد علاقة رياضية للتنبؤ ببداية صدأ حديد التسليح اعتماداً على المعطيات المحدودة من هذه التجارب. إن النتائج المخبرية أكدت أن عامل الانتشار لأيون الكلوريد في الخرسانة يتأثر فعلياً بنسبة الماء للإسمنت والأجواء المحيطة، وهامشياً بكمية الإسمنت ولايتأثر بتركيز نسبة الكلوريد على سطح الخرسانة. لقد حددت قيمة عامل الانتشار لأيون الكلوريد في الخرسانة يتأثر فعلياً بنسبة الماء للإسمنت والأجواء المحيطة، وهامشياً بكمية الإسمنت ولايتأثر بتركيز نسبة الكلوريد على سطح الخرسانة. لقد حددت قيمة عامل الانتشار بين ٨ و ١٧٠ × ١٠ سم<sup>×</sup>/ثانية لخلطات خرسانية وأجواء محيطة مختلفة. إن هذه القيم العالية لعامل الانتشار يمكن عزوها إلى رداءة نوعية الخرسانة وصعوبة الأجواء المحلية المحيطة. لقد تم وضع حجر الأساس لإمكانية تنبؤ بداية صداً حديد التسليح والتي قد أعطت نتائج مجدية نسبة إلى المعطيات المحدودة من هذه التجارب.

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## ABSTRACT

This investigation evaluates the chloride diffusion coefficient for concrete made with marginal limestone aggregates and Type V Portland cement commonly used in the Arabian Gulf. Parameters investigated are (1) water-cement ratio, (2) cement content, (3) surface chloride concentration, and (4) exposure condition. An attempt is made to arrive at a model for predicting the onset of corrosion using the limited test data. The experimental results revealed that the chloride diffusion coefficient is strongly influenced by water-cement ratio and exposure conditions and marginally by cement content but unaffected by the surface chloride concentration. The chloride diffusion coefficient values ranged from (8 to 170) × 10<sup>-8</sup> cm<sup>2</sup>/sec for different concrete mixes and exposure conditions. The high value of the chloride diffusion coefficient is mainly attributed to the low concrete quality and the local aggressive environmental conditions. The basis of the prediction model to corrosion initiation has been set and reasonable results are produced based on the limited experimental data.

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## **1. INTRODUCTION**

Concrete durability has been a major problem for concrete construction in the Arabian Gulf, where concrete structures appear to deteriorate at an extraordinary rate. One of the dominant causal factors of deterioration is recognized as the severe environmental and climatic conditions [1]. The concomitant presence of hot and humid climatic conditions with high seasonal temperature fluctuations and the high level of airborne and waterborne chlorides have in effect created a harsh, hostile environment for concrete construction leading to accelerated corrosion of reinforcement. Chloride-induced corrosion of reinforcement has been recognized as the single most dominant factor for widespread concrete deterioration.

Even for a concrete with negligible amount of initial chloride at the construction stage, the gradual built-up of the required level of chloride to initiate corrosion takes place through diffusion of chloride ions from external sources. Consequently, diffusion of chloride ions in concrete has been a major focus of research interest. Several researchers [2–19] in the past have established the applicability of Fick's second law of diffusion in estimating the chloride diffusion coefficient in concrete. The different parameters studied were the water–cement ratio, cement content, cement type, surface chloride concentration, and the use of additives to develop denser concrete. Most of these studies have been conducted with better concrete samples in temperate climatic conditions, findings from which may not be applicable to concrete using sulfate-resisting cement, with the aim of determining the values applicable to this region. The effect of varying water–cement ratio, cement content, surface chloride concentrations, and, importantly, exposure conditions were studied. Based on the limited laboratory test data, an attempt is also made to arrive at a model for predicting the onset of corrosion in the reinforcement of concrete structures.

## 2. EXPERIMENTAL PROGRAM

The flow chart shown in Figure 1 presents the overall testing program conducted for the different parameters to evaluate the chloride diffusion coefficient and to measure the initiation of corrosion. For evaluating the chloride diffusion coefficient in concrete, sixteen  $76 \times 46 \times 15$  (cm) concrete slabs were cast. The specimens were divided into two groups, one group to be tested under a controlled laboratory conditions and the other group earmarked for testing in the exposed, natural environment. For monitoring the initiation of corrosion, twelve concrete block specimens  $30 \times 15 \times h$  (cm) were cast with a central 12 mm diameter steel bar and kept in laboratory conditions. The depth of the concrete blocks h was varied to yield three different covers. The variations of water-cement ratio, cement content, surface chloride concentrations, and reinforcement cover are indicated in Figure 1.

## **3. SPECIMEN PREPARATION AND MEASUREMENTS**

ASTM C 150 Type V sulfate resisting cement, crushed limestone aggregates, and beach sand which are commonly used in local concrete construction were used for the preparation of all the specimens. The coarse-to-fine aggregate ratio was kept constant at 1.5 for all the mixes. The specimens were cast in three layers with proper compaction. Twenty-four hours after casting, the specimens were demoulded and cured in laboratory temperature of  $18-20^{\circ}$ C by covering with wet burlap for a period of two weeks. Following the curing period, one set of specimens was kept inside the controlled laboratory condition and the other was exposed to the outdoor environment, where summer temperature varies between 35 and 50°C.

The specimens were ponded with sodium chloride solutions eighty days after casting. Cement mortar dykes one inch high were constructed for the slabs and the blocks to facilitate the ponding. The joint between the surface of the specimens and the dyke was sealed with a silicon sealant and the inner side of the dyke was coated with an epoxy to prevent any leakage of the solution. Also, when one inch diameter cores were extracted periodically (every 35 days) from the slabs for chloride profile analysis, the holes so formed were also properly sealed to prevent leakage. The chloride content measurement was done using the spectrophotometric method [20].

The initiation of corrosion for the concrete block specimens were monitored at regular intervals using half cell potentials and corrosion current density measurements using the linear polarization technique. At the end of the monitoring period (300 days), all blocks were split open for visual inspection and the chloride content in concrete at the rebar level was determined.

### 3.1. Evaluation of Effective Diffusion Coefficient

The effective diffusion coefficient is evaluated using Crank's solution [21] for Fick's second law subject to the appropriate boundary conditions:

$$\frac{C_x - C_i}{C_s - C_i} = 1 - erf\left[\frac{x}{2\sqrt{D_e t}}\right]$$
(1)

where  $C_i$  is the initial chloride concentration,  $C_x$  is the chloride concentration at a depth x,  $C_s$  is the surface chloride concentration, t is the time elapsed,  $D_e$  is the effective diffusion coefficient and erf is the error function. The experimental measurements provide the data for t, x,  $C_i$ , and  $C_x$  in Equation (1). As it is practically impossible to measure the value of chloride concentration  $C_s$  at x = 0, Equation (1) contains two unknowns  $C_s$  and  $D_e$ . Therefore, the equation has to be solved using an iterative approach. An interactive optimization computer program was used to analyze the experimental data. The program takes into account the initial chloride concentration and determines the best fit for the experimental data over the specified range of  $C_s$  and  $D_e$ . The final value of  $D_e$  is selected based on the minimum sum of square of errors (SSE) for an appropriate value of  $C_s$ . The effective diffusion coefficient  $D_e$  and the surface chloride concentration  $C_s$  are collectively listed in Table 1.



Figure 1. Flow Chart of Overall Testing Program.

The use of Fick's laws of diffusion to concrete underlies one basic assumption, that no binding takes place between the ingressing chloride ions and the tri-calcium in concrete. The applicability of this assumption to concrete made with Type I Portland cement has been verified [22]. In this investigation, Type V Portland cement has been used which has much lesser tri-calcium aluminate than Type I and consequently much lesser binding with chloride ions.

#### **3.2. Prediction of Time to Corrosion Initiation**

An attempt is made to use the limited test data to arrive at a model for predicting the time to initiation of corrosion in concrete. From the statistical analysis of the experimental data, a regression model for the effective diffusion coefficient  $D_e$  is obtained as a function of two important variables, namely water-cement ratio (w/c) and cement content ( $C_c$ ).

$$D_e = \left[ 82.7 - 425.9(w/c) + 568.4(w/c)^2 + 4.3(C_c)^{-6} \right] \times 10^{-8}.$$
 (2)

In Equation (2),  $C_c$  is normalized with respect to a value of 350 kg/m<sup>3</sup>, *i.e.*  $C_c$  is the ratio of the cement content in kg/m<sup>3</sup> divided by 350 kg/m<sup>3</sup>. The  $D_e$  values obtained from Equation (2) are plotted *versus* the measured experimental values as shown in Figure 2, which indicate a fairly close fit.

Using the solution of Fick's second law (Equation 1) for the appropriate boundary conditions and the regressed model for  $D_e$  from Equation (2), the initiation time t for corrosion can be predicted assuming a threshold value  $C_x$  of chloride concentration. The threshold chloride concentration  $C_x$  can be taken as 0.35% by weight of cement as verified by the measurement of chloride content at the rebar level of the concrete blocks which showed initiation of corrosion, and as recommended by various code stipulations [23].

A prior knowledge of surface chloride concentration  $C_s$  is needed for a prediction of initiation time. As a concrete in general is subjected to a variable degree of chloride exposure through seasonal and diurnal fluctuations of environmental and climatic condition,  $C_s$  varies. However, for practical purposes, assuming a constant value of  $C_s$  which would be a fair representative of the actual insitu condition, will yield good estimation. For this prediction, the values obtained from the optimization run were used.

	Cl	$CC = 350 \text{ kg/m}^3$						
W/C		Indoors			Outdoors			
		. D <sub>e</sub> *	<i>C</i> <sub>s</sub> **	SSE	<i>D</i> <sub>e</sub> *	<i>C</i> <sub>s</sub> **	SSE	
0.40	4%	8.03	0.32	0.0015	24.45	0.362	0.0031	
	8%	8.52	0.56	0.0019	27.54	0.748	0.0262	
0.55	4%	_	_	-	78.00	0.950	0.0263	
	8%	28.10	0.760	0.0165	73.50	1.250	0.0703	
0.70	4%	68.45	0.580	0.0055	165.0	0.832	0.0411	
	8%	66.30	0.815	0.0178	170.3	1.950	0.1204	
W/C		Indoors						
	Cl		CC = 300  kg/m	n <sup>3</sup>		CC = 400  kg/m	n <sup>3</sup>	
0.40	4%	10.32	0.49	0.0022	7.63	0.38	0.0036	
0.55	4%	33.70	0.54	0.0100	16.46	0.49	0.0010	
	•							

Table 1. Effective Diffusion Coefficient  $D_e$  and Surface Chloride Concentration  $C_s$ .

 $* \times 10^8$  cm<sup>2</sup>/sec

\*\* % by weight of concrete

## 4. RESULTS AND DISCUSSION

The chloride diffusion measurements in concrete for the different parameters investigated are too voluminous to be presented in this paper. However, samples of these measurements are shown in Figures 3-6, taken at 175 days, which represent the effect of water-cement ratio (w/c), indoor/outdoor exposure condition, cement content (cc) and surface chloride concentration, respectively.



Figure 2. Experimental versus Calculated Value of D<sub>e</sub>.



Figure 3. Effect of w/c Ratio on Chloride Diffusion: (cc = 350 kg/m<sup>3</sup>, 175 days, 8% chloride, indoors).

## 4.1. Effect of W/C Ratio

The chloride diffusion and hence the diffusion coefficient are strongly influenced by the w/c ratio as indicated in Table 1 and shown in Figure 7. The increase in diffusion with w/c ratio is in agreement with previously reported data [4, 8, 9, 19, 24]. From their study on OPC mortar, Gjorv and Vennesland [24] reported the effect of higher w/c ratio to be confined only to top surfaces (*i.e.* upto a depth less than 10 mm) and the diffusion in the interior portions to be affected by other factors



Figure 4. Effect of Exposure Conditions on Chloride Diffusion: (cc = 350 kg/m<sup>3</sup>, w/c = 0.4, 8% chloride, 175 days).



Figure 5. Effect of Cement Content on Chloride Diffusion: (w/c = 0.55, 175 days, 4% chloride, indoors).

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like chloride binding. However, for SRPC concrete, it is seen that the effect of w/c on diffusion is not limited to the surface layers only but also occurs in the interior layers. It can be observed from Figure 3 that with an increase in the w/c from 0.4 to 0.7, after 175 days of exposure, the chloride penetration depth increased from 4 cm to 10 cm.



Figure 6. Effect of Exposure Solution Concentration on Chloride Diffusion: ( $cc = 350 \text{ kg/m}^3$ , w/c = 0.4, 175 days, outdoors).



Figure 7. Variation of Effective Diffusion Coefficient with w/c Ratio.

### 4.2. Effect of Exposure Condition

The effect of the two exposure conditions, *viz.* indoors and outdoors, on chloride diffusion is shown in Figure 4. The indoor exposure of controlled laboratory conditions was at an average temperature of 19°C. The outdoor exposure period ranged from August to March, during which the ambient temperature ranged from 8 to 50°C. For all concrete mixes, there was a marked difference in the values of the diffusion coefficients between the two exposures. The diffusion coefficient for specimens exposed to outdoor conditions was 3 times that of the value for indoor specimens made with a w/c ratio of 0.40 ( $8.03 \times 10^{-8}$  to 24.45  $\times 10^{-8}$  cm<sup>2</sup>/sec). However, the difference between the outdoor and the indoor is reduced for higher w/c ratios. The sharp difference in the values of diffusion coefficient between the two exposure conditions may be attributed mainly to the effect of temperature on the kinetics of diffusion and the associated factors.

An increase in temperature will lead to a decrease in the viscosity of the pore water in concrete, through which the diffusion takes place, and consequently an increase in ionic diffusion. Moreover, at higher temperatures, for example  $55^{\circ}$ C and above, C<sub>3</sub>A does not react with chloride ions to form stable chloro-aluminates [25]. Thus, the assumption of negligible reaction taking place can be valid in this case. The possible formation of microcracks at the aggregate paste interface from the high temperature fluctuations due to the thermal incompatibility between the aggregates and the cement paste [26, 27] in the exposure condition is another contributing factor. The minimal binding effects together with the development of microcracks would lead to a higher chloride diffusion in concrete exposed to harsh outdoor environment as observed in this research.

#### 4.3. Effect of Cement Content

The decrease in chloride diffusion with an increase in cement content is attributed to the development of a denser concrete with smaller porosity and pore radius. However, at low water-cement ratio, such effect becomes marginal as shown in Figures 5 and 7.

#### 4.4. Effect of Exposure to Chloride Concentration

The values of surface concentrations obtained from the optimization run are shown in Table 1. As expected, in all the cases, the ingress of chloride ions increased considerably for higher exposure chloride concentration as shown in Figure 6. However, the diffusion coefficients were almost unchanged for the same w/c ratio, irrespective of the ponded solution concentration. For example, in the case of w/c = 0.40, the diffusion coefficients in concrete ponded with 4 and 8% chloride solutions are  $8.03 \times 10^{-8}$  cm<sup>2</sup>/sec and  $8.52 \times 10^{-8}$  cm<sup>2</sup>/sec, respectively.

Even though the diffusion coefficients are independent of the exposure chloride concentration, the effect of high exposure concentrations is detrimental from the durability point of view, as the chloride ion built-up is proportional to the surface chloride concentration,  $C_s$ .

#### 4.5. Prediction of Corrosion Initiation

The corrosion potentials and the corrosion current density values for some typical concrete block specimens of 3.8 cm cover are shown as representatives in Figures 8 and 9. The time to initiation of corrosion for the various specimens as inferred from the half cell and the corrosion density measurements are collectively presented in Table 2. To examine the actual state of the rebars after 300 days of exposure to chlorides, all the block specimens were split open. Table 2 presents the chloride contents at rebar level measured after 300 days of exposure.

Based on the diffusion coefficients calculated from Equation (2), the surface chloride concentration  $C_s$  values obtained from the optimization run, and for an assumed chloride threshold value of 0.35% by weight of cement (approximately 0.052% by weight of concrete) at the reinforcement level, the time to initiation of corrosion can be predicted using Equation (1). The predicted time to corrosion initiation for the different w/c, exposure to chloride solutions and rebar covers are given in Table 3. A comparison of the predicted and the actual time to corrosion initiation gives a reasonable correlation as indicated in Table 3. The variation between the predicted and the actual initiation times does not follow a definite trend. However, if experiments are conducted considering a more wider range for the two variables considered, *viz*. w/c ratio and cement content, a model of more general applicability could be developed using the same approach.

		4% S	olution	8% Solution		
W/C	Cover (cm)	Chloride Content*	Corrosion Initiation (Days)	Chloride Content*	Corrosion Initiation (Days)	
0.40	10.2	0.003	Nil	0.003	Nil	
	7.6	0.003	Nil	0.003	Nil	
	3.8	0.0031	Nil	0.051	280	
0.55	10.2	0.0048	Nil	0.012	Nil	
	7.6	0.010	Nil	0.048	285	
	3.8	0.125	200	0.244	120	

Table 2.	Chloride	Contents	Measured	at	Rebar	Leve	l
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\* % by weight of concrete

	Table 3. Pr	redicted Time to Co	prosion Initiation.			
		Cover to Reinforcement (cm)				
W/C	Chloride	10.2	7.6	3.8		
		Time to Corrosion Initiation in Days (Years)				
0.4	4%	3887 (10.6)	2186 (6.0)	547 (1.50)		
	8%	2700 (7.3)	1519 (4.20)	380 (1.04) 280 (0.787)*		
0.55	4%	1015 (2.8)	571 (1.6)	143 (0.39) 200 (0.548)*		
	8%	709 (1.9)	399 (1.10) 285 (0.781)*	100 (0.27) 120 (0.329)*		

\*Experimental results

## **5. CONCLUSIONS**

- 1. The chloride diffusion coefficient for sulfate-resisting Portland cement concrete under the local construction environment of the Arabian Gulf appears to be greater than the values reported thus far in literature. The outdoor exposure condition significantly increases the diffusion coefficient.
- 2. Diffusion coefficient  $D_e$  is found to be strongly influenced by the water-cement ratio. The value of  $D_e$  increases with increasing w/c ratio but decreases with an increase in the cement content. The influence of cement content is more pronounced for higher w/c ratios.
- 3. The chloride diffusion coefficient appears to be unaffected by the surface chloride concentration. However, as the amount of chlorides within the concrete is directly proportional to the surface concentration, higher surface concentration is detrimental from the viewpoint of durability as it will lead to an earlier initiation of corrosion.
- 4. The expression obtained for  $D_e$  of chloride ion in concrete was based on the limited variables considered in this study. Tests have to be conducted considering a much wider range of the different parameters, which would help in arriving at a more general expression of  $D_e$  which could be of great practical use.

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Figure 8b. Corrosion Potentials: (w/c = 0.4, 4% chloride, cover = 3.8 cm).

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Figure 9b. Corrosion Potentials: (w/c = 0.4, 8% chloride, cover = 3.8 cm).



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