CHLORIDE DIFFUSION AND CORROSION ACTIVITY: MODELLING THE EFFECT OF BINDER TYPE AND CONTENT

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ABSTRACT

This paper describes the theoretical basis for the ingress of chlorides into concrete. It is recognised that there are differences in the overall ingress rates in fully saturated and partially saturated concrete. Furthermore, precise universal models for concrete exposed to a chloride environment are not possible. Thus, it is argued that it is necessary to determine the relative resistance of each concrete type to chloride ingress to be able to estimate the durable lifespan of a particular structure. Two simple methods are described to enable this. The first uses Fick's 1st Law to provide an estimate of exposure period required induce corrosion providing that a reliable coefficient of diffusion is available and is applicable to new construction. The second method uses an existing chloride concentration profile to estimate residual durability and is, therefore, applicable to existing structures. A comparison is made of the improvement to chloride resistance that can be achieved by incorporating different pozzolanic binders and also effect these have on corrosion activity. Both conventional binary blends as well as relatively novel multi-component binders are considered and it is shown that the latter group can provide greatly enhanced chloride resistance.

KEYWORDS

Chloride ingress, binary/multi-component binders, pulverized-fuel ash, ground granulated blastfurnace slag, metakaolin, silica fume, corrosion, modelling, estimation of durability.

INTRODUCTION

Chloride ingress into concrete structures continues to be a problem in many countries from temperate northern Europe to the Middle East. Although the sources of chlorides may be different, the end result is the same, ie destructive corrosion of reinforcement. In the Arabian Gulf region these problems can be especially acute as the high average ambient temperature results in higher rates of chloride flux than occur in winter highway conditions in northern climes. However, the pozzolanic activity of binders, such as pulverized-fuel ash (PFA), ground granulated blastfurnace slag (GGBS), silica fume (SF) and metakaolin (MK) is also stimulated by higher ambient temperatures and can provide an effective way of retarding chloride ingress

Modelling the process of chloride ingress into concrete would appear to be relatively straightforward, since the basic physics of ionic transportation through a semi-permeable material are well established. However, the physical and chemical heterogeneity of binders and concrete, together with the fact that the hydrates are not inert and can chemically immobilise chlorides, means that simple models have been difficult to establish.

This paper draws on extensive studies into chloride ingress and reinforcement corrosion that have been carried out at the Concrete Technology Unit at Dundee University and compares the relative advantages of using pozzolanic binders in binary and multi-component combinations with PC. The theoretical basis for modelling chloride ingress is then used to provide methods for estimating chloride build-up in concrete. The effect of different binders on chloride-induced corrosion has also been considered.

THEORETICAL BASIS FOR CHLORIDE INGRESS INTO CONCRETE

Chloride transportation into concrete involves both ionic diffusion and capillary absorption, and is likely that a mixed mode will occur depending upon the prevailing exposure conditions, and the pore saturation and microstructure of the concrete (Concrete Society, 1996). The process is further complicated by the inhomogeneity of concrete, changes in concrete properties with increasing maturity and any chemical interaction of chloride ions and the binder.

Most methods of modelling and measuring chloride ingress simply rely on pure diffusion with concrete assumed to be sufficiently water-saturated. The parameter normally adopted to reflect resistance to chloride ingress in concrete is the diffusion coefficient (D), which may be modified to account for chloride binding and absorption in unsaturated concrete. The coefficient of chloride diffusion can also be used for specifying durable life and several diffusion-based design methods have been proposed (eg. Tuutti, 1981, Browne, 1982, Dhir et al, 1991).

Diffusion in Saturated Concrete

Fickian models of diffusion are used by many researchers to describe chloride ion transportation in saturated concrete. In these diffusion models, the driving force is the chloride concentration difference in the pore water. There are a number of different methods of determining D, based either on Fick's First or Second Laws of Diffusion as shown below;

Fick's First Law $$J = -D\frac{dC}{dx}$$

Fick's Second Law $$\frac{\delta C}{\delta t} = D\frac{\delta^2 C}{\delta x^2}$$

Where: J = Flux ($mole/m^2/s$)
D = Coefficient of diffusion (m^2/s)
dc/dx = Concentration gradient
t = Time (s)
C = Concentration (g/m^3)
x = Distance (m)

Modified Reaction/Diffusion

The application of Fick's laws of diffusion assumes that the substrate is a homogeneous, isotropic and inert medium. The assumptions, therefore, imply that the diffusion properties at any point in concrete would be the same in all directions and do not change with time. In reality, these conditions do not exist in concrete which is composed of relatively "unstable" hydrates which interact both physically and chemically with the diffusing species. Different binders have different capabilities in relation to taking up and binding chlorides. Cements containing a high C_3A level and cements containing MK, PFA and GGBS, which are high in reactive alumina, can slow down the rate of chloride ingress in concrete considerably.

Models based on non-Fickian behaviour have been developed which account for linear and non-linear chloride binding (Sergi et al, 1992, Nilsson et al, 1994, Andrade et al, 1996 and Papadakis et al, 1996). However, several mathematical models are proposed and there appears to be a lot of ambiguity on the appropriate diffusion coefficient that should be used.

Absorption/Diffusion in Unsaturated Concrete

The above methods of determining D assume that concrete is water saturated. However, in most chloride-bearing exposure conditions, periodic wetting and drying of the structure takes place. In these situations, chlorides are likely to enter the concrete under both absorption (capillary suction) when the concrete is dry, and by diffusion once the pores become filled. Cycles of chloride solution absorption and water evaporation produce a cumulation of chloride at the concrete surface. Chloride diffusion rates in these exposures have been shown to be greater than those under saturated conditions (Petersson, 1993; Henderson, 1997).

Several numerical models to determine diffusion coefficients, taking into account sorption effects and changing surface concentration, have been reported (Bentz et al, 1996, Tang & Nilsson, 1992). These models use a direct finite difference implementation of Fick's second law and corrections for the effect of initial sorption, changing surface concentration with time and chloride reaction with the hydrates. However, these methods are complex and further work is needed in this area to verify their validity.

EXPERIMENTAL DETAILS

Materials

The binder materials used, as a direct replacement to PC, were metakaolin (MK), silica fume (SF), pulverized-fuel ash (PFA) and granulated blastfurnace slag (GGBS). A series of combinations for binary and multi-component binders, which are permitted within the current draft standard for common European cements: DD ENV 197-1(1995), were considered as shown in Table 1. Potentially these mixes should allow engineers, through careful selection and combination, to exploit the benefits associated with the use of blended cements in concrete. Table 2 summarises the main characteristics of the binder combination used in this study.

The aggregates were crushed, siliceous gravel, in two single-size fractions of 20 -10mm and 10 - 5mm. The fine aggregate was a natural sand of Zone M to BS 882 (1992). All aggregates were dried in laboratory air prior to their use in concrete production.

Mix Proportions and Test Methods

The mix proportions used for reference PC concretes are given in Table 3. For binary and multi-component binder concretes, the free water and binder contents were fixed, as for the corresponding PC concrete, but the fine aggregate contents were adjusted to maintain the yield. For all tests, concrete specimens were cured in water at 20°C, prior to strength testing or exposure to the accelerated chloride test environment. Compressive strength tests were performed on 100mm cube specimens in accordance with BS 1881: Part 116 (1983). Chloride diffusion (D) was measured using a standard two compartment test cell developed at the University of Dundee (Dhir et al,1990).

Table 1. Various combinations for (a) binary and (b) multi-component binders (% mass)

MIX	PC	PFA	GGBS	SF	MK
(a) Binary Mix Group					
	90 - 75	-	-	-	5 - 25
	95 - 80	-	-	5 - 20	-
(b) Multi-Component Mix Group					
A	100	-	-	-	-
B	66.5	33.5	-	-	-
C	66.5	30.0	-	3.5	-
D	66.5	30.0	-	-	3.5
E	50.0	-	50.0	-	-
F	50.0	-	50.0	5.0	-
G	50.0	-	45.0	-	5.0

Table 2. Key binder characteristics.

BINDER CHARACTERISTIC		PC	SF	MK	PFA	GGBS
Specific Surface Area, m^2/kg		380	15,750	3475	450	510
Main Oxides, % wt	SiO_2	21.4	95.3	55.1	51.1	34.2
	Al_2O_3	4.7	0.65	40.4	24.9	13.9
	Fe_2O_3	2.7	0.28	0.6	9.0	0.6
	CaO	65.2	0.27	0.03	1.50	41.6
	MgO	1.0	0.40	0.40	1.40	-
	SO_3	2.9	0.25	-	0.70	-
	LOI	0.9	-	1.2	5.7	0.9
	K_2O	0.64	0.77	-	3.6	-
	Na_2O	0.13	0.26	0.01	1.60	0.50
Bogue Composition, % wt	C_3S	57				
	C_2S	18				
	C_3A	7.8				
	C_4AF	8.2				

Table 3. Mix proportions for PC control concrete.

DESIGN STRENGTH, N/mm^2	CONSTITUENT MATERIALS, kg/m^3					W/C ratio
	Free water	PC	Aggregate			
			Sand	10 mm	20 mm	
25	185	235	700	400	800	0.79
35	185	285	730	400	800	0.65
40	185	310	710	400	800	0.60
50	185	355	670	400	800	0.52
60	185	410	625	400	800	0.45

EFFECT OF BINDER TYPE ON CHLORIDE INGRESS

Binary Binders

The results show that with PC/MK and PC/SF binders, based on direct replacement by mass, coefficients of chloride diffusion (D) were considerably lower than corresponding PC concrete. The effect of different replacement levels on D of 355 kg/m^3 binder content concrete, together with the corresponding 28 day strength is shown in Figure 1. These are typical plots of the data obtained with all binder contents considered in the study. It is clear that the use of MK and SF lead to a reduction in D and this increased with material replacement level.

Across the range of binder contents, reductions of approximately 2 to 3 times, compared to the PC concrete were observed for PC/MK10, with larger reductions as the MK level increased to 25% replacement for PC. Similar reductions were obtained for SF concretes. The results also indicate that at a given replacement level, the SF concrete had a slightly higher D value than corresponding MK concretes. This may reflect the higher alumina content of MK, which contributes to greater chloride binding and may reduce the rates of chloride ion diffusion.

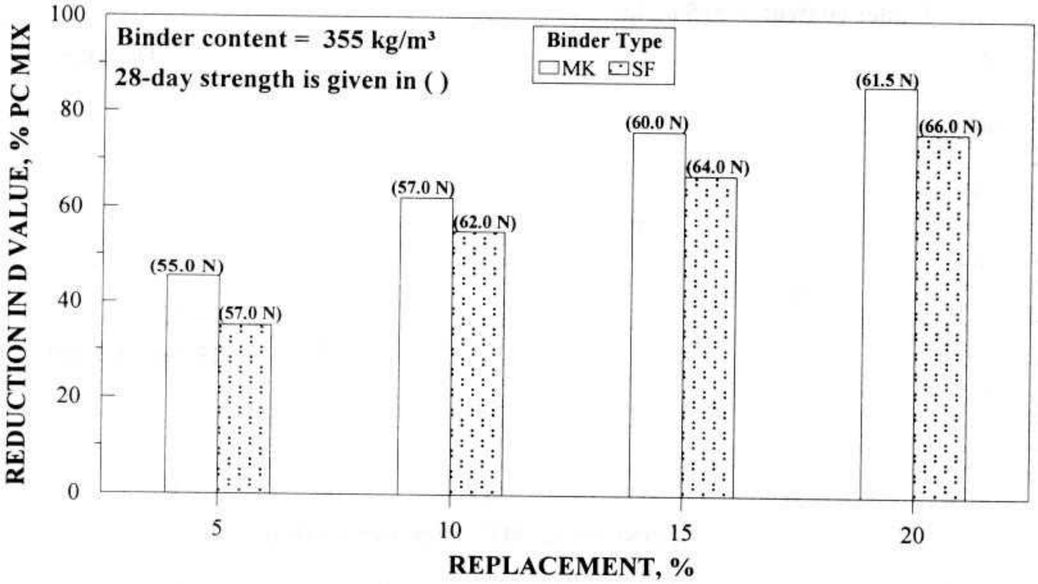

Fig. 1. Effect of MK and SF binders on chloride diffusion into concrete.

Multi-Component Binders

Despite the improvements achieved with the use of MK and SF in concrete, the demands placed upon modern concrete construction are becoming greater and so there is a need for higher performance concrete. For example, much improved chloride resistance for concrete exposed to aggressive chloride environments. It is important to appreciate that, in general, chloride durability of concrete is largely a function of the cement paste and its interfaces.

Given this, a series of multi-component binders (Table 1b) were tested with constant total binder and water contents. Results obtained from chloride diffusion and compressive strength tests on these concretes are given in Figure 2. This illustrates how some binder combinations may provide improved chloride resistance and poorer strength performance in some cases. It is clear from these results that chloride resistance of concrete cannot not be solely judged by strength since the chemical binding capacity of the binders also has an important role.

The results show that the use of multi-component binders greatly improves the chloride resistance properties of concrete when compared with PC concrete. For example, concrete mixes F and G give the best performance of the GGBS concretes, with relatively similar strength and low chloride diffusion. On the other hand, the chloride diffusion rates for PFA mixes (C and D) were improved, but the strength of these mixes were slightly reduced. From these results, it should be possible, depending on the exposure conditions, to select the most suitable binder combination for optimum performance. Bearing in mind that the method of specification is normally centred around a required design strength, it is likely that on this basis, the majority of multi-component binder concretes will provide further enhanced durability performance compared to those of PC concrete.

These results show that there is a need to change that method of specification of concrete where chloride durability is an overriding requirement since strength can neither account for chloride binding capacity nor any effects that pozzolanic binder produce in refining the pore structure and paste interfacial zones.

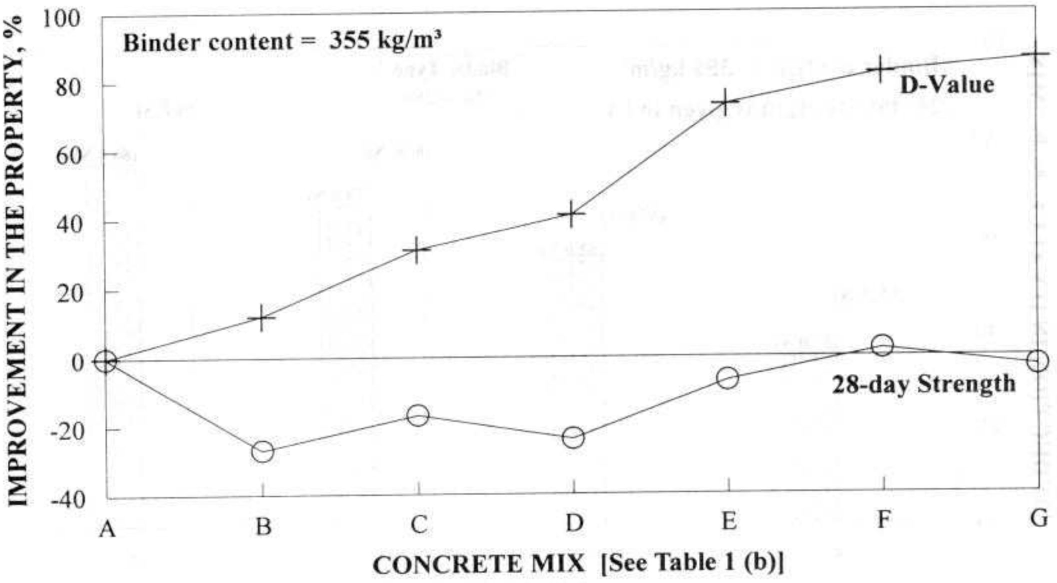

Fig. 2. Influence of combining PFA and GGBS with other binder materials on strength and chloride diffusion properties of concrete.

EFFECT OF WATER/BINDER RATIO ON CHLORIDE INGRESS

Binary Binders

In order to examine the effect of water/binder (w/b) ratio on chloride diffusion rates, coefficient of chloride diffusion against w/b ratio curves were generated, using the data obtained above. A typical example of this relationship for concrete made with 10% MK and SF binders is shown in Figure 3 (a). The results indicate that D values for PC/MK and PC/SF concrete mixes were considerably lower than the corresponding PC concretes, at all w/b ratios, with PC/MK binders being slightly more effective. At 0.52 w/b ratio, D for concrete containing 10% MK was 5.0 x 10-11 m^2/sec compared to 6.2 x 10-11 m^2/sec for concrete made with 10% SF. Similar effects were observed for the other replacement levels at all w/b ratios. This was expected as MK has a high alumina content, which contributes to greater chloride binding and may cause a reduction in chloride diffusion rates.

Multi-Component Binders

The chloride diffusion test results from multi-blend concrete mixes containing SF and MK with PC/PFA and PC/GGBS binders are plotted against w/b ratio to examine their effect. The results demonstrate the benefits of using multi-component binder mixes in reducing chloride diffusion rates and subsequently extending concrete performance. Figure 3(b) shows, as a typical example, that considerable improvements can be achieved with the use multi-component binders in concrete production, at a given w/b ratio.

Clearly, such relationships can be useful to establish, for a given chloride-bearing environment, the adjustment to w/b ratio necessary to take advantage of the beneficial effects of the binary or multi-component binders and integrated with mix proportioning methods to achieve a required durability performance.

EFFECT OF CONCRETE ON RATES OF CHLORIDE-INDUCED CORROSION

In addition to its influences on the rate of chloride ingress into concrete, the binder type also appears to have an influence on corrosion activity. This is illustrated in Figure 4, which gives the results from a study considering corrosion activity in both PC and PC/PFA concrete. This indicates that some binders after a given period of exposure have better resistance to the onset of corrosion than others. Clearly this is related to the fact that for PFA concrete, chloride build-up is slower, such that after a given exposure period the contamination level is lower.

Work has shown that once corrosion has initiated, for a given level of contamination, the rate may differ considerably between different concrete and concrete types. The literature suggests that there are a number of different factors which may contribute to these effects. Some of the more important parameters are:

- Binder/hydrate/reaction product composition.
- Chloride and oxygen availability and ease of movement to the corrosion site.
- Concrete microstructure.
- Concrete resistivity.
- Pore fluid chemistry.

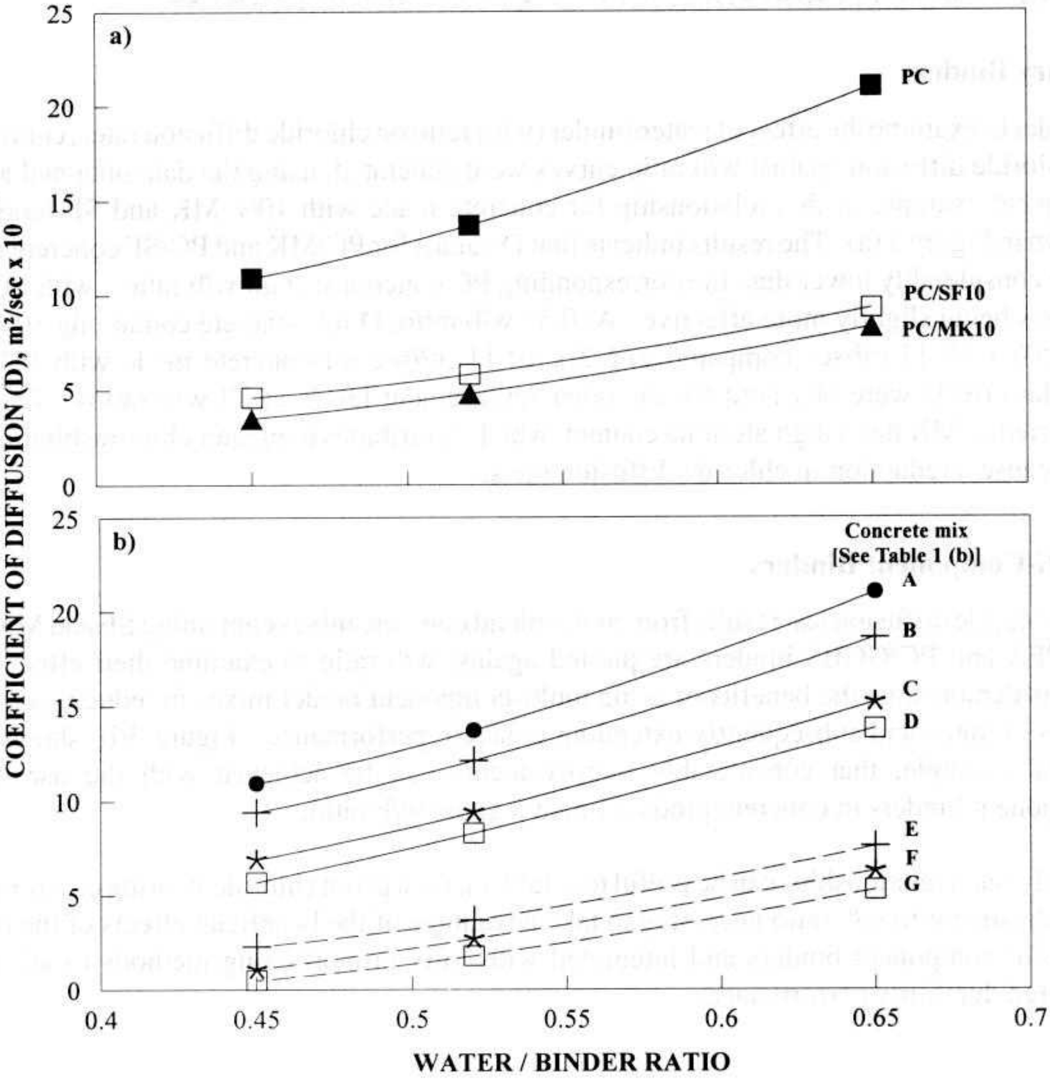

Fig. 3. Effect of water/binder ratio on (a) binary and (b) multi-component binder concrete chloride diffusion coefficient.

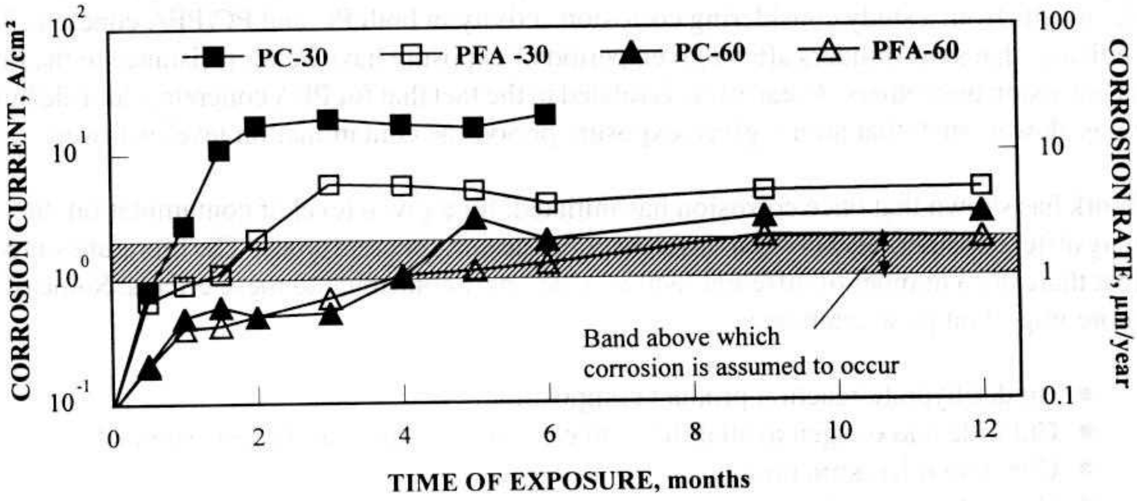

Fig. 4. Effect of binder type and concrete grade on time to initiate of corrosion and the rate at which corrosion intensity develops thereafter.

Some of these are closely related and interdependent and the magnitude of influence of each on the process would be very difficult to establish. However, the position becomes clearer if consideration is given to the relationship between chloride concentration (water-soluble) and corrosion current (rate). This is shown in Figure 5 and illustrates that, as may be expected, with increasing level of chloride contamination, the rate of corrosion increases and on initial inspection, there appears to be no direct relations between the two parameters. However, by considering the coefficient of chloride diffusion for the concretes tested, it is possible to rationalise the data and group it into bands.

The addition of D to the relationship is not arbitrary (although the ranges included are), since it is a measure of the overall rate of transportation of chloride through concrete and is influenced by the concrete microstructure and chemistry. In cases where D is low, the rate of corrosion tends to be low and where this is high, the rate tends to be higher.

This result is of practical significance, since the measurement of D will provide an indication of what the likely risk and rate of corrosion may be. A tentative classification has been devised and is shown in Table 4. This, based on the relationship observed in Figure 5, links the coefficient of chloride diffusion and chloride concentration to the corrosion intensity.

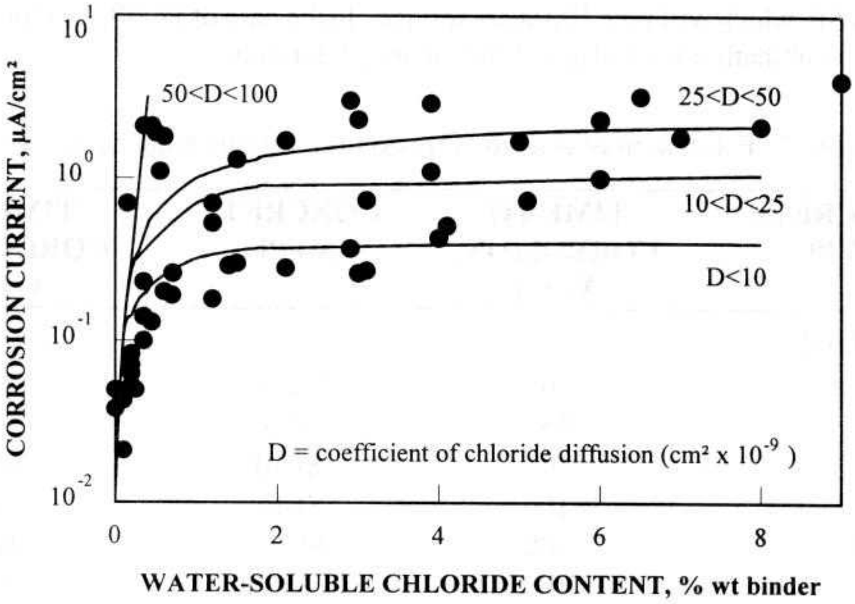

Fig. 5. Relationship between corrosion intensity and D.

Table 4. Estimation of corrosion risk using chloride content and D.

D: cm^2/s x 10^{-9}	**CORROSION INTENSITY**		
	Low*	**Medium***	**High***
	Water-Soluble Chloride Content, wt% binder		
<20	1.0	N/A	N/A
20-50	0.3	0.5	2.0
>50	0.2	0.4	0.6

*Low = 0.1μA/cm^2 *Medium = 1.μA/cm^2 *High = 10.0μA/cm^2

ESTIMATING CHLORIDE DURABILITY OF CONCRETE

Estimations Based on Coefficient of Chloride Diffusion

In order to provide practical information on the effectiveness of the various binders considered, it is possible through the use of estimation models such as that of Dhir et al (1991) to establish the period of service that may be expected, prior to the initiation of corrosion. This is based around Fick's diffusion laws and coefficient of chloride diffusion and has been developed for use by engineers to allow service life to be built into the design process .

This method can be illustrated by considering a typical structural concrete, with a range of different binders, used in a chloride exposure with w/c ratio of 0.5, well cured, a water-soluble chloride threshold level of 0.2% weight of binder, a cover depth of 50 mm and an external concentration of 0.5M in the environment. Based on the laboratory measured D values, Table 5 has been generated and provides an indication of the period required to initiate corrosion. The results indicate that for MK and SF that progressive increases in service life were achievable with increasing level and approximately 100 years can be obtained when a 15% replacement is used. It is apparent for the multi-blend concretes that the service life depends on the combination considered. For PC/GGBS, the best combinations appear to be with MK and SF, which will give 100 years service. In the case of PC/PFA as the main binder components, combination with MK and SF enhanced durability.

Table 5. Estimation of time to corrosion for various binder concretes

CONCRETE TYPE	TIME TO CORROSION, Years	CONCRETE TYPE	TIME TO CORROSION, Years
Binary Blends			
PC	30	PFA33	40
MK5	45	SF5	40
MK10	70	SF10	65
MK15	>100	SF15	80
MK20	>100	SF 20	>100
MK25	>100	GGBS50	55
Multi Blends			
PC/PFA30/SF3.5	50	PC/GGBS45/SF5	95
PC/PFA30/MK3.5	50	PC/GGBS45/MK5	>100

Estimations Based on Historical Data

A new approach to modelling chloride ingress which can potentially overcome the problem of unknown local microclimate, construction effects etc. has been recently put forward by Dhir et al (1998). The mathematical models developed are primarily based on the assumption that the total chloride content profile within a semi-infinite medium can be expressed as an exponential decay function of the Boltzmann variable. The mathematical basis of the method has been given in detail by Dhir et al (1998) and should be consulted for further information.

In summary, the steps to determine the likely future development of a chloride ingress profile are as follows:

Step 1 Plot the historical total chloride content against √t and obtain S_n which is the slope. It is satisfactory in laboratory accelerated tests to use data for as short an exposure period as 1 month to obtain reliable estimates of S_n. However, it is recommended that at least 3 month chloride penetration profiles are obtained. That is particularly so for highly chloride resistant concrete.

Step 2 Using the following equation: $k = \frac{2}{S_{cn}}$

Calculate k, which is the decay rate of the chloride profile.

Step 3 Using the following equation: $C_n = e^{-k\varphi}$

Calculate Cn, which is the total chloride content at any given time and where φ is the Boltzmann variable (mm/√month).

Step 4 Plot the C_n against φ to determine the future development of the chloride profile, substituting any required cover depth and time increments to determine φ.

Example Application of the Historical Data Method

Chloride penetration data for two grade 40 mixes, Mix A: a normal Portland cement concrete and Mix B: a 50 % PFA concrete water-cured at 20°C for 28 days before exposure to a 5.0M chloride environment at 20°C was obtained (Dhir et al, 1993). Figure 6 shows the S_{cn}/root time curve for this data, from which k can be calculated. Using the calculated values for k, Figure 7 has been plotted and is compared with the actual chloride penetration data obtained. The corresponding fit between the data is good and supports the validity of the method.

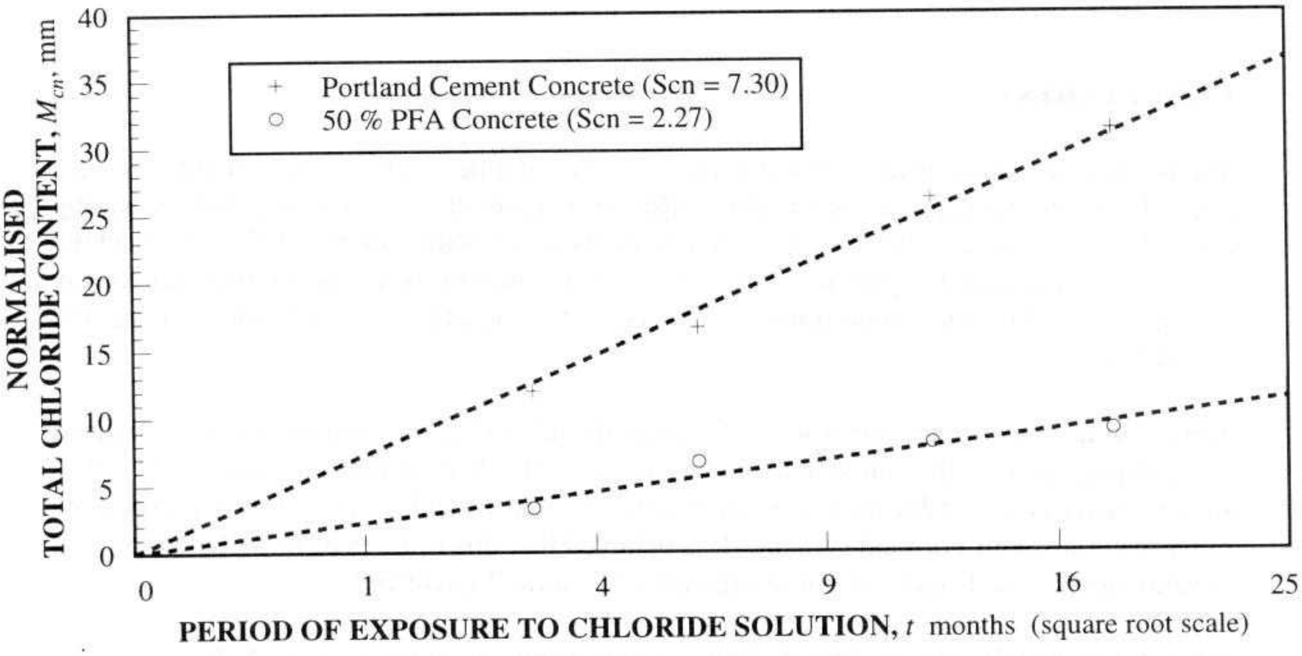

Fig. 6. Normalised total chloride content for two grade 40 concrete immersed in a 5M NaCl solution at 20^0C.

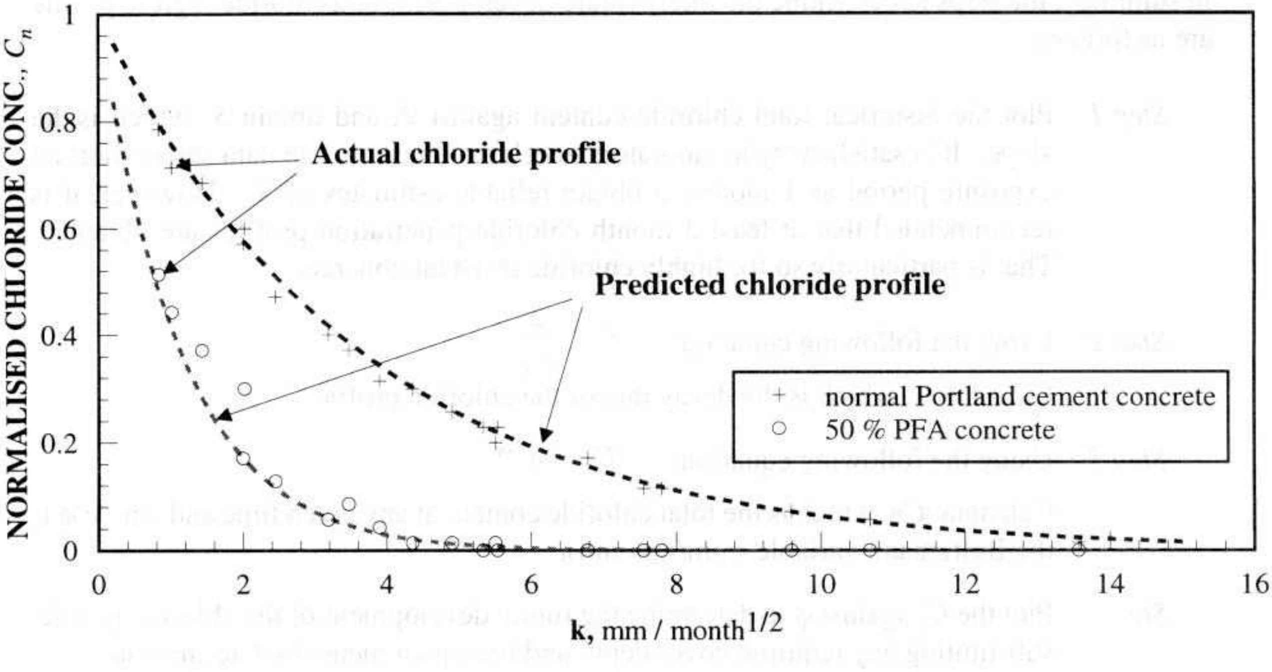

Fig. 7. Normalised chloride content profiles for two grade 40 concrete immersed in a 5M NaCl solution at 20^{0}C.

The main drawback to this method is that it will only be accurate if future exposure conditions do not change greatly from those from which the historical data was gathered. It is, therefore, suggested that on a practical basis data should be gathered after several years to ensure that the full range of seasonal effects are experienced. In areas where there is considerable variation in the climate conditions it may be necessary to base future estimations of durability on 10 year historical data for it to be valid.

CONCLUSIONS

The mechanisms of chloride transport in concrete are complicated since they can involve both ionic diffusion and capillary absorption. However, methods of modelling and measuring chloride ingress are usually based on Fickian models of pure diffusion which do not reflect the conditions experienced in practice. These models can however, be modified, in a limited way to take account of such conditions by correcting for the effects of chloride binding and absorption.

The chloride ingress test results showed that significantly lower diffusion rates can be obtained using binary and multi-component binders in concrete than in that containing PC. It is suggested that considerable improvements can be obtained through combination of two or more other materials with Portland cement. It is believed that this route is of importance towards developments in and achievement of high chloride durability concrete.

It was found that the rate of chloride-induced reinforcement corrosion was related to the rate at which chlorides can be recharged to the corrosion site. In turn, this is related to the coefficient of chloride diffusion, thus concrete with a low D value will tend to also have a relatively slower rate of corrosion. A classification for corrosion rate based on chloride concentration and D value has been proposed. However, it should be noted that in terms of overall serviceability the corrosion propagation period even in low D concrete will be much shorter than the corrosion initiation period. Consequently, it is recommended that in design for durability, key importance is given to achieving the minimum practical chloride transportation rate to the reinforcing steel.

Two different ways of estimating chloride ingress were described. The first method is shown to be applicable to new construction where diffusion is the dominant chloride transportation process but requires the D value to be determined in the laboratory. The effect of different binders on the time to initiate corrosion have been compared. It was found that the estimated time to corrosion for a typical structural grade PC concrete could be increase from around 25 years to 100 years by using a multi-component mix of PC, MK and SF binders. The second method is applicable to existing structures and provides a more accurate estimation of residual durability by using the chloride profile that has built-up from the particular exposure environment. The particular advantage of this is that will take account of the wide number of variables that affect chloride ingress.

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