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Reinforcing steel corrosion in high-volume fly ash concrete

S. Qian, D. Qu, G.P. Gu and N. Bouzoubaa

The corrosion of reinforcing steel in high-volume fly ash (HVFA) concrete slabs was investigated. Chloride ions were introduced into the reinforced concrete slab specimens through a natural diffusion and migration process, that is, immersion in a ponding solution of 3.4 percent sodium chloride (NaCl). Two HVFA concretes specimens with ASTM Class C and Class F fly ash were made with a water-to-cementitious materials ratio (w/cm) of 0.32; besides, three control concretes specimens with a w/c of 0.32, 0.43 and 0.55 were also investigated. The concrete cover depths to the steel reinforcing bars ranged from 13 mm to 76 mm. The corrosion of the reinforcing steel bars was evaluated by means of half-cell potential, linear polarisation and AC impedance techniques. The results indicated that the concrete incorporating Class C fly ash showed the best performance with respect to chloride-induced reinforcing steel corrosion, followed by the control concrete with the w/c of 0.32 and concrete containing Class F fly ash; the control concretes with w/c of 0.43 and 0.55 showed the poorest performance.

Corrosion of rebars in reinforced concrete (RC) structures is recognised as a major problem in the maintenance of the structural integrity. The most important causes for initiation of corrosion of reinforcing steel are the ingress of chloride ions and carbon dioxide to the steel surface. Chloride ion causes local destruction of the passive film leading to localised corrosion. Carbon dioxide, on the other hand, reacts with the hydrated cement matrix, leading to a decrease in pH and subsequent loss of steel passivity and to corrosion initiation. The need to extend concrete durability has led to the use of several admixtures and modifications to the concrete composition. Addition of fly ash as an additive to concrete has become common practice in recent years[1]. The fly ash particles react with calcium hydroxide, producing cementitious products that strongly decrease concrete porosity[2-4]. This effect leads to an increase of the concrete resistivity and consequently to a decrease of the diffusivity coefficients of some elements, such as oxygen and chloride through the concrete[5-7]. It has been reported that a reduction of chloride diffusivity in immersed concrete to half its value was observed in concrete incorporating 50 percent of fly ash. Consequently, the corrosion process in the presence of chlorides was delayed. This behaviour was revealed in previous work on fly ash concrete immersed in sodium chloride solutions[8-10]. The use of 30 percent fly ash as partial substitution of cement has led to a significant increase of induction time and to a reduction of the corrosion rate by one order of magnitude[10].

In this paper, experimental investigations on the corrosion of reinforcing steel bars embedded in slabs containing high volume fly ash (HVFA) concrete were undertaken. The corrosion resistance of the rebars in slabs was then compared to a control mix of conventional portland concrete (PC) having the same w/cm ratio. Reinforcing steel bars were embedded in these slabs with different depths of concrete cover in order to investigate steel corrosion induced by chlorides. The chloride profile was assessed using acid soluble chloride analysis technique to examine the effect of HVFA in reducing the ingress of chlorides into concrete. Tests of reinforcing steel bars embedded in PC with higher water/cement ratios (w/c = 0.43 and 0.55) were also included for comparison purpose.

Experimental programme

Concrete specimens

Air-entrained concrete specimens with and without high volumes of fly ash were made. Samples included two HVFA concrete mixtures made with 58 percent Class F (N1) and Class C (N2) fly ash with a w/cm of 0.32, and three concrete

Table 1: Mixture proportions and compressive strength of the concrete

Mix no	*Concrete description*	*Fly ash content, percent*	*w/cm*	*w/c*	*Quantities, kg/m³* Water	Cement	Fly ash	FA	CA	SP	*AEA, ml/m³*	*f'c (28-days), MPa*
N1	HVFA-F	58	0.32	0.76	119	156	217	751	1124	4.0	403	36.4
N2	HVFA-C	58	0.32	0.76	119	156	217	747	1120	5.0	403	55.8
N3	Control-0.32	0	0.32	0.32	120	376	0	769	1150	9.1	457	61.1
N4	Control-0.43	0	0.43	0.43	160	372	0	723	1085	0.5	111	37.7
N5	Control-0.55	0	0.55	0.55	165	300	0	736	1099	0	60	31.0

FA - Fine aggregate, CA - Coarse aggregate, SP - Superplasticiser, AEA - Air-entraining agent, Control-0.32 denotes control concrete with a w/c ratio of 0.32

Table 2: Guidelines for half-cell potential data interpretation

E versus $Cu/CuSO_4$ (ASTM C876), mV	*E versus Hg/Hg_2Cl_2, mV (in this study)*	*Corrosion activity*
>-200	> -128	90 percent probability of no corrosion
-200 > E > -350	-128 > E > -278	Corrosion is uncertain
< -350	< -278	90 percent probability of corrosion

mixtures of conventional portland concrete with w/c = 0.32, 0.43 and 0.55 (N3, N4, and N5, respectively). A multi-component, synthetic-resin, air-entraining admixture (AEA) was used in all the concrete mixtures. The compositions of the concrete mixtures are listed in *Table* 1. The geometrical configuration of the concrete slab was 833 × 600 × 153 mm as shown in *Fig* 1.

The embedded reinforcing bars were 15 mm in diameter black steel, 470 mm in length and an exposed surface of 2.21×10^4 mm^2 achieved by coating the two ends of the reinforcement bars with epoxy (the extra surface area of the ribs on the reinforcement was not counted). Four pairs of reinforcing steel bars were embedded in each slab with concrete covers of 13, 25, 51 and 76 mm, *Fig* 1. The slabs were fabricated in April 1997 and cured under wet burlap for seven days followed by exposure to air in the laboratory for 28 days. They were then ponded with a 3.4 percent NaCl solution for 5.3 years (duration of the experiment). The properties of the concrete slabs have been reported elsewhere[11, 12].

Corrosion testing techniques

There are several non-destructive techniques available for the investigation of corrosion in reinforced concrete. In order to get a reliable assessment of the corrosion of reinforcing steel, three corrosion evaluation techniques were used in this study. They were half-cell potential, linear polarisation and AC impedance methods.

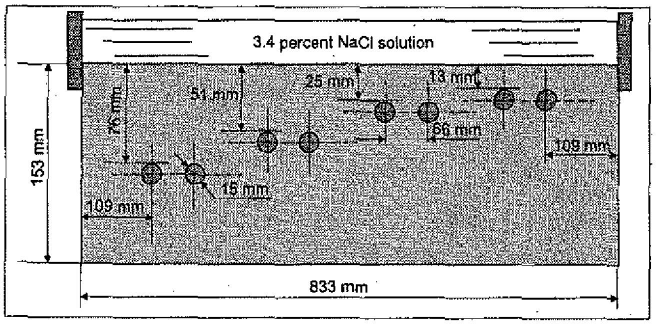


Fig 1 A schematic plot showing the portions of reinforcing steel bars in test slabs

Half-cell potential method

The half-cell potential method has been widely used because of its simplicity and cost effectiveness[13,14]. This method allows the evaluation of the probability of corrosion activity through the measurement of the potential difference between a standard portable reference electrode and the reinforcing steel. The data analysis guidelines described in ASTM C876-99 provide general principles for the evaluation of the probability of corrosion of reinforcing steel in concrete structures.

A saturated Hg/Hg_2Cl_2 electrode (SCE) was used to measure the potential of the reinforcing steel, because the ponded water on top of the concrete samples contained chloride ions. The guideline of half-cell potential specified in ASTM C876 is versus $Cu/CuSO_4$ electrode (CSE). The standard potential of saturated $Cu/CuSO_4$ and Hg/Hg_2Cl_2 is 318 mV and 246 mV versus that of the standard hydrogen electrode (SHE), respectively. The difference between these two reference electrodes is 72 mV. The guidelines described in ASTM C876 provide general principles for the evaluation of the reinforcing steel corrosion in concrete. The guidelines and their conversion values (versus SCE) are listed in *Table* 2.

Linear polarisation method

The linear polarisation technique was used to determine the polarisation resistance and the corrosion rate of reinforcing steel bars embedded in concrete. The potential of the steel electrode was scanned at a slow rate of 0.1 to 0.05 mV/s. The tests were initiated at 20 mV below the corrosion potential, E_{corr}, and terminated at 20 mV above it, while recording the polarisation current (I). These tests were conducted with one of the following potentiostats, EG&G 273 or 6310, Gamry CMS100, and Solartron 1286 or 1287. The polarisation resistance, R_P, of the reinforcing steel is defined as the slope of a potential-current density plot at the corrosion potential, E_{corr}, as follows:

$$R_p = \left(\frac{\Delta V}{\Delta I}\right)_{E_{corr}} \qquad ...(1)$$

Considering the correction of potential drop due to concrete resistance (iR drop correction), R_P is:

$$R_p = \left(\frac{\Delta V - \Delta I \cdot R_c}{\Delta I}\right)_{E_{corr}} \qquad ...(2)$$

where,

ΔV and ΔI = the applied potential and current response, respectively,

R_c = the concrete resistance between the reference electrode and the surface of reinforcing bar, which can be obtained by AC impedance technique.

The corrosion current density is calculated from the Stern-Geary equation[15]:

$$I_{corr} = \frac{B}{R_p} \qquad \text{...(3)}$$

and

$$B = \frac{b_a b_c}{2.303(b_a + b_c)} \qquad \text{...(4)}$$

where,

B = the so-called "Stern-Geary constant" that can be determined from the b_a and b_c,

b_a and b_c = the Tafel slopes for the anodic and cathodic reactions, respectively.

A value of 26 mV and 52 mV is often used in the calculation for the bare steel in the active and passive stages, respectively[16]. For simplicity, a value of 26 mV was used to calculate all the corrosion rates in this paper since the corrosion current is inversely proportional to R_P. The criteria for estimating the reinforcing steel corrosion are listed in *Table* 3[17].

AC impedance method

The measurement of AC impedance spectroscopy provides information on the electrical resistivity and the dielectrical properties of the concrete cover, the corrosion rate and the mechanism of reaction on the steel/concrete interface. This technique is frequently used in the laboratory to study the corrosion of steel in concrete. Experimental investigations have shown a close relationship between the corrosion rate determined by weight loss and the values calculated from AC impedance measurements[18,19].

The AC impedance measurements were performed by a solartron SI 1287 Electrochemical Interface coupled with SI 1260 HF Frequency Response Analyzer (FRA) and controlled by a PC computer with Zplot and Corr-ware software. A small sinusoidal voltage signal of 5 mV was applied over the range of frequencies 100 kHz to 0.0005 Hz. The experimental results were fitted by Zview software based on the equivalent circuit after which the polarisation resistance and the corrosion rate were calculated.

Results and discussion

Half-cell potential of reinforcing steel

Half-cell potentials were measured on the reinforcing steel bars embedded in the conventional concrete with different w/c ratios and HVFA concrete. *Fig* 2 shows the half-cell potential (average of two steel reinforcing bars with same depth of concrete cover) change with increase of the concrete cover depth measured after 5.3 years ponding with 3.4 percent NaCl solution. The results indicated that the reinforcing steel bars exhibited high corrosion rates in slab N5 (control concrete with w/c of 0.55) regardless of the concrete cover depth. In slab N4 (control concrete with w/c of 0.43) the reinforcing steel bars with concrete cover depths of 13 mm and 25 mm also displayed high corrosion rates; the only exception was those bars with 75 mm concrete cover. In all other slabs, the reinforcing bars were not corroded when the concrete cover depth was more than 25 mm, however the half-cell potential readings indicated corrosion of all reinforcing steel bars with a 13 mm concrete cover depth, except for slab N2. As expected concrete covers (>13 mm) can significantly delay the ingress of chlorides into the concrete/steel interface region.

Table 3: Criteria for estimating reinforcement corrosion conditions

Corrosion rate, µA/cm²	*Extent of corrosion*
$I_{corr} < 0.1$	P : passive condition
$0.1 < I_{corr} < 0.5$	L : low to moderate corrosion
$0.5 < I_{corr} < 1.0$	M : moderate to high corrosion
$I_{corr} > 1.0$	H: high corrosion

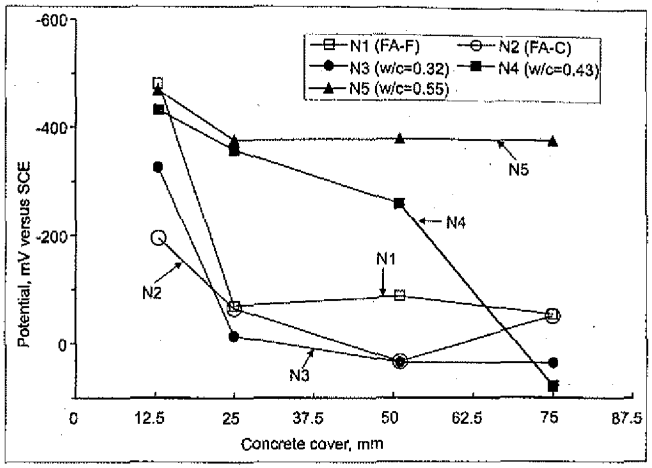


Fig 2 Half-cell potential measurements of reinforcing steel bars embedded in concrete slabs after 5.3 years of ponding with 3.4 percent NaCl solution

The half-cell potential readings for the 13 mm concrete cover depth, varied widely from slab to slab, *Fig* 3, which

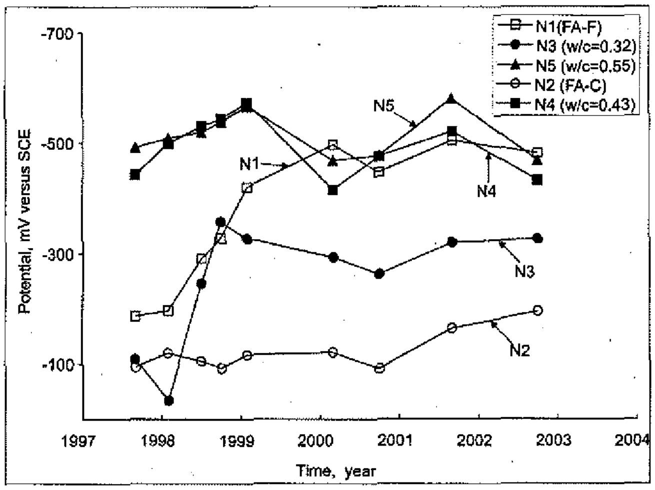


Fig 3 Half-cell potential measurements of reinforcing steel bars embedded in concrete slabs with 13 mm concrete cover

Table 4: Probability of reinforcing steel corrosion evaluated for different concrete mixes and differing concrete cover depths from half-cell potential measurements

Slab no	*Concrete description*	*Concrete cover depth, mm* *13*	*13*	*25*	*25*	*51*	*51*	*76*	*76*
N2	HVFA-C	H	L	L	L	L	L	L	L
N3	Control-0.32	H	U	L	L	L	L	L	L
N1	HVFA-F	H	H	U	L	L	L	L	L
N4	Control-0.43	H	H	H	H	H	U	L	L
N5	Control-0.55	H	H	H	H	H	H	H	H

Note: L: 90 percent probability of no corrosion; U: Uncertain state of corrosion; H: 90 percent probability of corrosion

Table 5: Corrosion state of reinforcing steel evaluated for different concrete mixes and differing concrete cover depths from linear polarisation

Slab no	*Concrete description*	*Concrete cover depth, mm* *13*	*13*	*25*	*25*	*51*	*51*	*76*	*76*
N2	HVFA-C	L	P	P	P	P	P	P	P
N3	Control-0.32	L	L	P	P	P	P	P	P
N1	HVFA-F	L	M	P	P	P	P	P	P
N4	Control-0.43	H	H	H	H	H	H	P	P
N5	Control-0.55	H	H	H	H	H	H	H	H

Note: P: Passive; L: Low; M: Moderate; H: High

made it possible to compare the performance of different types of concretes. The potentials for concrete slabs N1, N4 and N5 shifted into the most negative range in 2002, while N2 (HVFA Class C fly ash) maintained the most positive values followed by slab N3.

Table 4 lists the detailed probability of corrosion for all reinforcing steel bars embedded in the concrete slabs (including control concrete slabs with different w/c ratios) with different thicknesses of concrete covers evaluated after 5.3 years of ponding in 3.4 percent NaCl solution by half-cell potential technique according to the ASTM C876. It clearly shows that the concrete slab N2, that is, the concrete slab with HVFA Class C fly ash had the best performance, followed by slab N3 (control concrete w/c = 0.32) and slab N1 with HVFA Class F fly ash. The concrete slabs N4 and N5 (control concrete with w/c ratio of 0.43 and 0.55, respectively) had the poorest performance.

Corrosion current measurements by linear polarisation

Fig 4 shows the corrosion current density obtained by the linear polarisation technique. The corrosion current densities (average of two steel reinforcing bars) are plotted versus increasing cover depth for the concrete slabs containing high volume fly ash and the control slab (w/c = 0.32). When the thickness of concrete cover is 25 mm or higher, the corrosion current densities of reinforcing steel bars were all lower than 0.1 $\mu A/cm^2$ indicating that they were still in the passive condition. However, the difference in the corrosion current densities on the reinforcing steel bars with 13 mm concrete cover were quite significant. It is clearly shown that the concrete slab N2 (Class C fly ash) had the best performance, followed by slabs N3 (control w/c = 0.32) and N1 (Class F fly ash). The results from slabs N4 and N5 are not shown in *Figs* 4 and 5 since their current densities on most reinforcing bars were very high, *Table* 5.

Fig 5 illustrates the changes in corrosion current densities with time on the reinforcing steel bars with a 13 mm concrete cover. These measurements were carried out for up to 5.3 years of ponding in 3.4 percent NaCl solution. It can be seen that the corrosion current density started to increase from 1998 and reached the highest, then reduced in 2002 for the slab N3 (control slab). In contrast, the current density for slab N2 remained the lowest.

Table 5 summarises the corrosion state of reinforcing steel bars embedded in all the slabs evaluated by the linear polarisation method after 5.3 years of ponding in 3.4 percent NaCl solution. It shows clearly that the concrete slab N2 had the best performance followed by slab N3 and N1. As expected, the slabs N4 and N5 had the worst performance.

Corrosion current measurements by AC impedance

To interpret the AC impedance spectra, an equivalent circuit fitting procedure is commonly used. The complexity of the reinforced concrete (RC) system makes this approach difficult. Sometimes, different models have to be tested to obtain the best fit and also to have physical meaning for the metal/concrete interface[20-22].

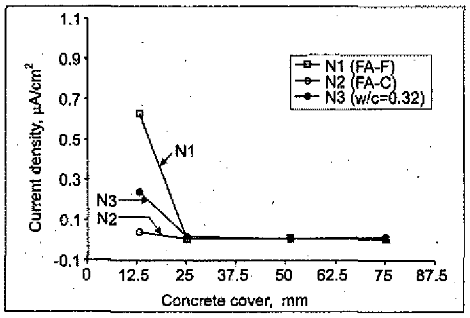


Fig 4 Corrosion current densities obtained by linear polarisation technique on reinforcing steel bars embedded in concrete slabs (measurements after 5.3 years of ponding with 3.4 percent NaCl solution)

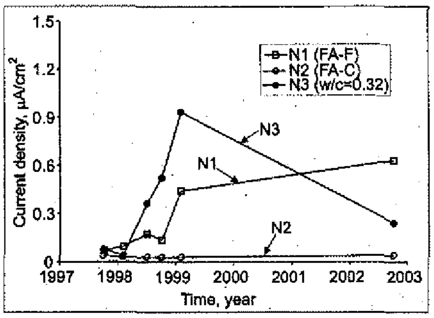
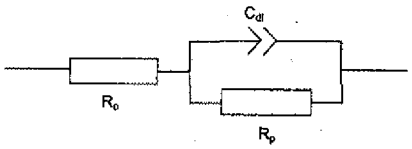


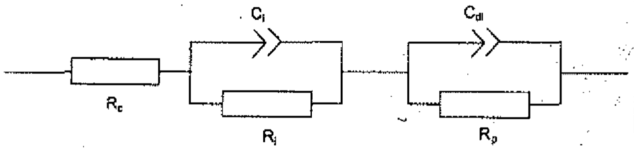
Fig 5 Corrosion current densities obtained by linear polarisation technique on reinforcing steel bars embedded in concrete slabs with 13 mm concrete cover

It is generally accepted that the physical model of the steel/concrete interface consists of a layer of iron oxides and hydroxides film in the passive stage and an interfacial film adjoined to the concrete matrix[21-23]. Many equivalent circuits have been proposed to describe the different stages of the steel/concrete corrosion process including active, and passive corrosion processes involving diffusion control, passive film formation and macro-cell corrosion etc[22,24-26].

The corrosion current for reinforcing steel in concrete slabs was



(a) simple equivalent circuit



(b) modified equivalent circuit

Fig 6 Equivalent circuit used in AC impedance fitting process

Table 6: Corrosion state of reinforcing steel evaluated for different concrete mixes and concrete cover depths from AC impedance*

Slab no	*Concrete description*	*Concrete cover depth, mm*							
		13	*13*	*25*	*25*	*51*	*51*	*76*	*76*
N2	HVFA-C	P	P	P	P	P	P	P	P
N3	Control-0.32	L	L	P	P	P	P	P	P
N1	HVFA-F	L	L	P	P	P	P	P	P
N4	Control-0.43	H	H	M	H	L	L	P	P
N5	Control-0.55	H	H	H	H	H	H	H	H

Note: P: Passive; L: Low; M: Moderate; H: High

obtained using the AC impedance technique. The values of the polarisation resistance, R_p, and the concrete resistance, R_c, were obtained by the "best-fit" of the experimental impedance spectra using one of the equivalent circuits shown in *Fig* 6. Two types of equivalent circuits were used in this work. A simple equivalent circuit (circuit (a)) was used in most of the analyses. In this equivalent circuit, a pure resistor, represent ohmic resistance, R_c, is in series with a parallel combination of a resistor (polarisation resistance, R_p) and a frequency dependent double layer capacitor, C_{dl}. The latter, also called a constant phase element (CPE) is introduced to account for a depressed semicircle on complex plot[27,28]. A modified equivalent circuit (circuit (b)) was also used in this study. This equivalent circuit consisted of a resistor, R_c, in series with two parallel combinations of a resistor and a CPE, R_i with C_i and R_P with C_{dl}. These RC parameters are defined as follows:

- R_c: the concrete resistance
- R_i and C_i: steel/concrete interface film resistance and capacitance
- R_p and C_{dl}: steel polarisation resistance and double-layer capacitance.

The complex-plane plot of AC impedance measurement data (symbols) and fitting results (solid line) for reinforcing steel bars embedded in the concrete slab N2 is shown in *Fig* 7. The fitted value of R_p represents the overall condition of the steel surface corrosion and does not identify the passive or active corrosion areas even though the latter has the major contribution to the measured Rp[29]. The value of R_p was used to calculate the corrosion rate based on the Stern-Geary relation (equation 3) and a "B" value of 26 mV was applied. The value of R_c was used to correct the *iR* drop in the linear polarisation measurement.

The relation of corrosion current density of reinforcing steel bars versus concrete cover (after 5.3 years ponding with chloride solution) are plotted in *Fig* 8. The reinforcing steel bars with a 25 mm concrete cover or thicker in all slabs had a very small corrosion current density (< 0.1 μA/cm^2). Higher corrosion current densities were observed for most concrete slabs with 13 mm concrete cover except the concrete slab N2 in which the current density remained very low. The results for slabs N4 and N5 were not shown in this figure since their values were too larger for this figure, *Table* 6.

The corrosion rates evaluated by the AC impedance method were also analysed using the criteria listed in *Table* 2, as shown in *Table* 6. It is clearly seen that the slab N2 with Class C fly ash had the best performance followed by the slabs N1 (Class F fly ash) and N3 (the control concrete w/c = 0.32). The difference in the current density between slabs N1 and N3 was very small. The worst performances were those of slabs N4 and N5.

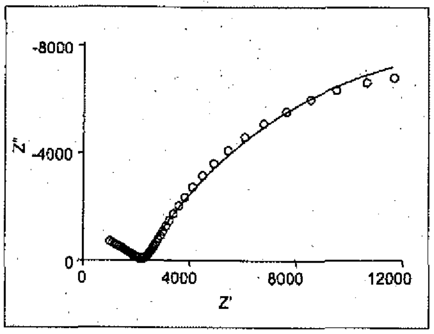


Fig 7 A complex-plane plot of AC impedance for reinforcing steel bars embedded in concrete slab N2; the symbols represent the experimental data and the line the fitted results

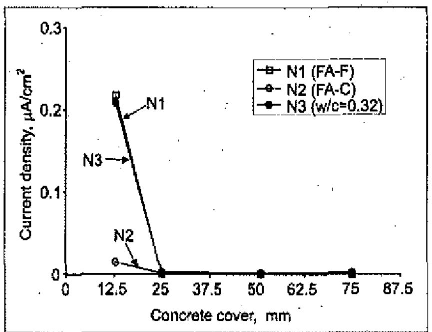


Fig 8 Corrosion current densities obtained by AC impedance technique on reinforcing steel bars embedded in concrete slabs (measurement after 5.3 years ponding with 3.4 percent NaCl solution)

Table 7: Overall performance ranking with regard to corrosion of reinforcing steel bars in concrete slabs

Slab no	*concrete description*	*Raking by different techniques*		
		Half-cell	*LP*	*AC*
N2	HVFA-C	1	1	1
N3	Control-0.32	2	2	2
N1	HVFA-F	3	3	2
N4	Control-0.43	4	4	4
N5	Control-0.55	5	5	5

Overall performance ranking of concrete slabs

The performance of the different concrete slabs tested using the half-cell potential and linear polarisation AC impedance techniques are listed in the *Table* 7. It can be seen that they are in very good agreement. The slab N2 (HVFA Class C fly ash) had the best performance followed by the control slab N3 (control concrete w/c = 0.32) and slab N1 with respect to the reinforcing steel corrosion. The slabs N4 and N5 (control concretes with w/c = 0.43 and 0.55, respectively) had the worst performance.

Conclusion

General

HVFA concretes (w/cm = 0.32) and control concrete (w/c = 0.32) performed excellently in the corrosion test reported. The reinforcing steel bars with a cover of 25 mm or greater were in a passive state after 5.3 years of exposure in 3.4 percent *NaCl* solutions. As most specifications for structural concrete exposed to aggressive media specify a minimum cover of 37 mm, the test results obtained indicate no likelihood of corrosion of reinforcing steel in HVFA concrete with w/cm of 0.32 during the service life of concrete structure.

The data reported in this investigation together with chloride-ion profiles in concrete slabs can be used to develop prediction model for service life of concrete structures exposed to chloride-ions.

Specific

The corrosion of reinforcing steel bars embedded in the concrete slabs containing high volume fly ash was investigated by half-cell potential, linear polarisation and AC impedance techniques. The performance of these concrete slabs and concrete cover depths in delaying corrosion of reinforcing steel bars was evaluated after 5.3 years of ponding with a 3.4 percent NaCl solution. Their performance was compared to conventional portland cement concrete and concrete slabs with high w/c. The results obtained by these different techniques were in very good agreement.

The concrete slab N2 (containing high volumes of Class C fly ash) had the best performance. The corrosion of the reinforcing steel bars was initiated only on the steel bars with 13 mm concrete cover and the corrosion rate was lower than that obtained in the control slab N3 (w/c = 0.32).

The control slab N3 (control concrete, w/c = 0.32) and slab N1 (containing high volumes of Class F fly ash) also had a good performance. The corrosion of the reinforcing steel bars with a 13 mm concrete cover depth was in the moderate to high corrosion state, but the reinforcing steel bars with thicker concrete covers were still in the passive condition, according to the measured corrosion rates.

The corrosion of reinforcing steel bars embedded in portland cement concretes with high water/cement ratios (w/c = 0.43 and 0.55) was significant. Substantial corrosion of the reinforcing steel bars was found in slab N4 (w/c = 0.43) with a 50 mm cover and in slab N5 (w/c = 0.55), even with a 75 mm concrete cover.

Acknowledgement

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