



Influence of Binder Type on the Chloride Threshold Level for Steel Corrosion in Concrete

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Abstract

The present study concerns the influence of binder type on the chloride-induced corrosion being accompanied by the chloride threshold level (CTL), chloride transport and as their results the corrosion-free life. Two levels of cement content, 30% PFA and 65% GGBS concrete were employed. It was found that the most dominant factor to the CTL is the entrapped air void content at the steel-concrete interface, irrespective of the chloride binding capacity, binder type and acid neutralisation capacity of cement matrix. The CTL for lower interfacial air void contents was significantly increased up to 1.52% by weight of cement, whereas a same mix produced 0.35% for a higher level of voids. Because of a remarkable reduction in the diffusion for GGBS concrete, its time to corrosion ranges from 255 to 1,250 days, while the corrosion-free life for control varies from 20 to 199 days and for 30% PFA concrete from 200 to 331 days.

Keywords : CTL(Chloride threshold level), steel corrosion, air void, diffusion coefficient, binding capacity

1. Introduction

Approaches to raising the chloride threshold level (CTL) include enhancing the concrete properties by using a low water-cement ratio, a high cement content, replacement with pozzolanic material or use of a superplasticizer. Literature reviews show that an improved inhibition effect can be achieved when using pulverised fuel ash (PFA), ground granulated blastfurnace slag (GGBS), or silica fume (SF), which reduce the mobility of chloride ions in concrete by producing a denser cement matrix with fewer capillary pores due to the pozzolanic reaction. Also, there is a reduction in the electrical conductivity of concrete and the diffusivity of chloride, which may result in a reduction in corrosion¹⁻⁴⁾.

Another advantage of replacement with PFA or GGBS is an increase in chloride binding capacity⁵⁻⁷⁾. In particular, the binding capacity of GGBS has been reported to be 5 times higher than that of OPC. The high aluminate levels present in PFA and GGBS are most likely to be responsible for their binding characteristics. A reduction of the free chloride content due to PFA or GGBS may also reduce the corrosion risk.

The effect of concrete replacement on the CTL has received little coverage in the literature, in contrast to the

widely reported assessment of chloride diffusion, the reason being the much longer time to corrosion initiation for PFA or GGBS concrete, which is very resistant to chloride transport. Thomas and Matthews⁸⁾ reported 10-year experimental results on the performance of PFA concrete, including chloride transport, corrosion of embedded steel. Their research indicated that the CTL was reduced by an increase in the PFA ratio in binder, in spite of the high resistance to chloride penetration. They also showed that the time to corrosion can be prolonged and corrosion propagation restricted by the addition of PFA or GGBS. It seems that the CTL is strongly dependent on the environment, type of chloride source and mechanism of chloride transport.

The present study discusses the influence of PFA or GGBS concrete on the CTL as well as the transport of chloride and corrosion propagation. Also covered is the development of a method to measure the CTL and to quantify the effect of the steel-concrete interface environment on the CTL.

2. Experimental works

2.1 Concrete mixtures

Four types of concrete specimens were tested to measure the CTL in this study. Specimens were manufactured using

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British OPC, as well as blended OPC concretes plus 30% PFA and 65% GGBS. The oxide composition of these cement binders is given in Table 1.

The mixture proportion used in manufacturing the specimens was 350 and 282 kg/m³ OPC, 519 and 709 kg/m³ fine aggregate (grade M), and 1,355 and 1,307kg/m³ 10mm coarse aggregate for rich and lean mixes respectively. The water: binder ratio was 0.45 for both mixes. The mix proportion for 30% PFA and 65% GGBS concrete was 400 kg/m³ total binder, 680kg/m³ fine aggregate and 1,230 kg/m³ coarse aggregate, and the free water-cement ratio 0.4 for PFA and GGBS concrete. Details of the mix proportions and slump results for each mix are given in Table 2.

2.2 Specimen preparation

Concrete specimens were cast in a 150mm cube mould with a centrally located 20mm diameter mild steel bar. The non-homogeneous oxide film (millscale) on the steel bars was removed using sand blasting and dilute acid to expose the fresh steel surface. This gave a much smoother finish, with no risk of crevice corrosion. The ends of the steel bars were masked off using a cementitious coating, which was then covered with heat shrink insulation to avoid corrosion under the masking material. The various level of voids at the steel-concrete interface were produced by varying the degree of vibration.

Table 1 Oxide composition of OPC, PFA and GGBS concretes (%)

Oxide	OPC	PFA	GGBS
CaO	64.7	1.7	41.2
SiO ₂	20.7	48.7	34.2
Al ₂ O ₃	4.6	18.8	11.7
Fe ₂ O ₃	3.0	7.7	1.43
MgO	1.0	1.0	8.81
Na ₂ O	0.13	0.4	0.29
K ₂ O	0.65	1.9	0.31
Mn ₂ O ₃	-	-	0.3
TiO ₃	-	0.9	0.58
SO ₃	3.0	0.64	-
Ignition loss	1.30	1.52	1.29

Table 2 Mixture proportions for concrete specimens used in this study

Mix	W/C	G _{max} (mm)	Slump (mm)	Unit weight (kgf/m ³)				
				Cement	PFA	GGBS	Sand	Gravel
OPC (lean)	0.45	10	140	282	-	-	709	1,307
OPC (rich)	0.45	10	130	350	-	-	519	1,355
30% PFA	0.4	10	90	280	120	-	680	1,230
65% GGBS	0.4	10	80	140	-	260	680	1,230

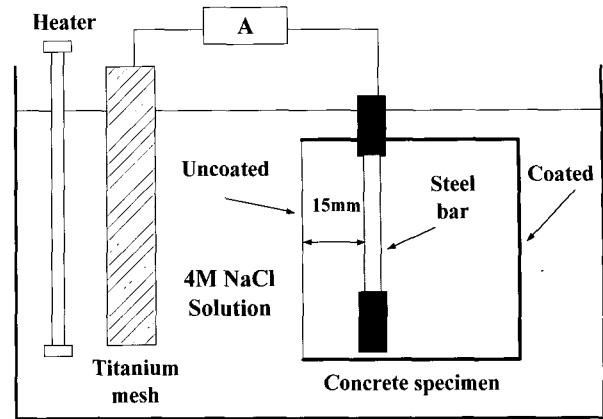


Fig. 1 Schematic of experimental set-up for CTL testing

After demoulding, the specimens were wrapped in polythene film and cured at 20±1°C for 4 weeks. Then specimens were sliced, parallel to the steel bar for exposure to the chloride solution, to produce a 15mm concrete cover depth. A barrier coating of epoxy resin was applied to seal the remaining cast surfaces to ensure diffusion from the one direction only. The specimens were initially saturated in a tank containing water for 1 week and then immersed in a 4M sodium chloride solution at 25°C. The end of the steel was electrically connected to an external cathode consisting of titanium mesh immersed in the aerated sodium chloride solution in the tank, as shown in Fig 1. During testing, the concentration of the solution was measured regularly to maintain a constant concentration. The level of water was also maintained at a constant level so that the specimens were totally immersed in the chloride solution.

2.3 Corrosion monitoring

The current flowing between the embedded steel and the titanium mesh was monitored every 6 hours by measuring the potential drop across a resistor (10kΩ) using a data-logger.

The measured galvanic current was plotted as anodic current against time. The value of galvanic current monitored can be varied with the concrete properties such as porosity, moisture content and resistivity of concrete that are influenced by replacement of highly grained minerals,

cement content and water-cement ratio. addition of admixtures such as calcium nitrite, which may influence the resistivity of concrete, may increase or decrease the galvanic currents in the passive and/or active state of the embedded steel. Corrosion initiation is indicated by a sharp increase in the current monitored between the steel and the cathode, showing an increase from a few micro amps to hundreds or thousands of micro amps.

An example of a plot of galvanic current per surface area of steel against time for an OPC sample is given in Fig 2, and shows a rise in current, indicating corrosion initiation. The specimens were subsequently removed from the tank and split so that the condition of the steel-concrete interface could be examined visually.

2.4 Visual examination

The voids were characterised and quantified under a petrographic microscopic using low magnification, combined with computer-based image analysis (Software: SCION IMAGE). Care was taken with lighting and in the choice of a video camera. The selection of a camera was

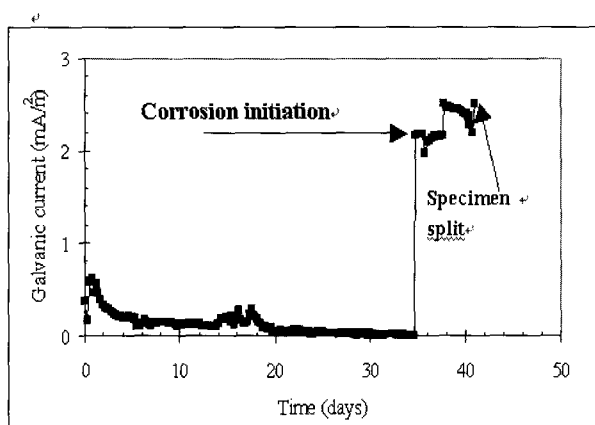
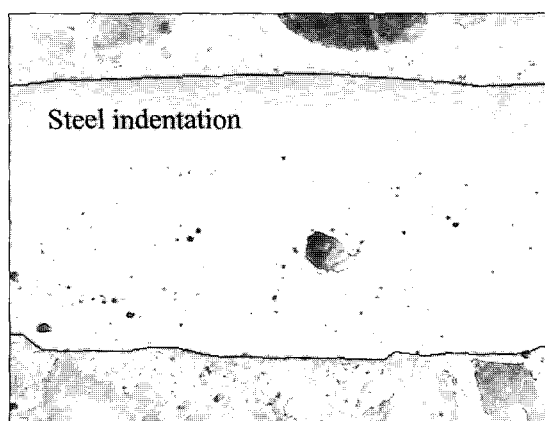


Fig. 2 An example of corrosion monitoring by galvanic current measurement between the anodic steel in concrete and an external cathodic titanium mesh



very important as it needed to have a very high depth of resolution to obtain an image of the curved surface. The lighting arrangement was adjusted to improve the accuracy of the method and to obtain a better image.

Following capture of the image, the concrete surface in contact with the steel was converted into white colour, and the voids into black colour, thus allowing the area percentage of voids to be calculated (Fig. 3). Voids more than 100µm in diameter were detected using this method. However, the rust stain generated, surrounding an entrapped air void, can be recognised as a void, because of its darkness (See Fig. 3). To avoid this, the rust in image was manually removed to turn white in binarised image. Hence, the void content under the rust stain may not be accounted, which may lead to a small error in calculating the void content at the steel-concrete interface.

2.5 Chloride profile

The chloride profiles were obtained from samples collected by grinding the concrete surface in 1mm depth increments within 2 hours of removing the concrete specimens from the tank. The chloride content of each sample was determined by acid soluble extraction in a nitric acid solution followed by potentiometric titration against silver nitrite.

This gave the chloride profile at the time the specimens were removed from the chloride solution tank; chloride diffuses into concrete when exposed to an external chloride source. The apparent chloride diffusion coefficient was determined on the specimens by fitting a solution to Fick's second law, for non-steady state diffusion in a semi-infinite medium.

Diffusion coefficient can be calculated by measuring the surface chloride and the chloride concentration at each depth of concrete to the depth of the steel. However, direct measurement of the surface chloride concentration is

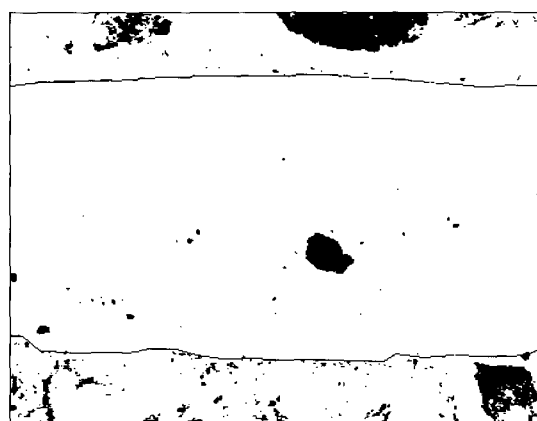


Fig. 3 An image of the steel-concrete interface (left) with air voids represented as dark pixels (right)

unreliable, because the surface chloride is the chloride content right at the exposed surface. The approach to obtain the surface chloride is to fit a curve to the chloride profile with the surface chloride and the diffusion coefficient as the independent variables. Also, in this study, background chloride content (i.e. from the mix ingredient) was neglected in analyzing the chloride profiles.

In the analysis of the chloride profile, the sum of the squared differences between the fitted (theoretical) and the actual data for the chloride concentration in each sample was minimised by adjusting the regressor variable, which in this case is the diffusion coefficient. Once a best fit was achieved, the diffusion coefficient and the surface chloride were then used to determine the CTL, i.e. the chloride content at the depth of the steel at the time of corrosion initiation. In this study, it was assumed that the difference in the diffusion coefficients between the time of chloride profiling and the time of corrosion initiation, because the gap is mostly less than 30 days, which is too small to change the diffusivity. Fig. 4 explains the approach for determining the CTL. Time t_1 represents the time of monitoring in the tank containing the chloride solution, while t_2 is the time to corrosion indicated by a significant rise in galvanic current.

3. Results

The experimental data obtained was analysed to give the time to corrosion initiation, the surface chloride content, the apparent diffusion coefficient and the CTL. The data was plotted as a function of the area of air voids at the steel-concrete interface.

3.1 Chloride threshold level

The CTL values obtained in specimens with different cement binders and degrees of vibration are illustrated in Fig. 5, which shows the effect of the air voids content at the steel-concrete interface. The curves for each binder take the form $y=ax^b$. Fig. 5 shows that the CTL varies inversely with the percentage of interfacial voids in concretes with the same mix. For all specimens, the CTL increased with a decrease in the interfacial void content. In particular, a sharp increase in the CTL was observed when the percentage of air voids decreased below 0.6%.

It was observed that the cement content in the mix also affects the CTL in OPC concrete. The CTL range for rich mixes was 0.48 to 1.52%, and for lean mixes was 0.35 to 1.4%. For a given air void content, the CTL for rich mixes was higher than for lean mixes by 0.2 to 0.5%. It is likely that this difference in CTL increases as the void content increases. In contrast, the CTL difference in relation to the cement content was negligible at low values of the air void

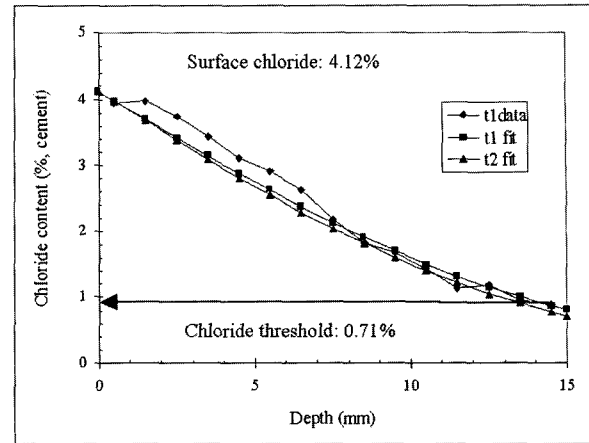


Fig. 4 Example showing how the CTL was determined from the chloride profile data

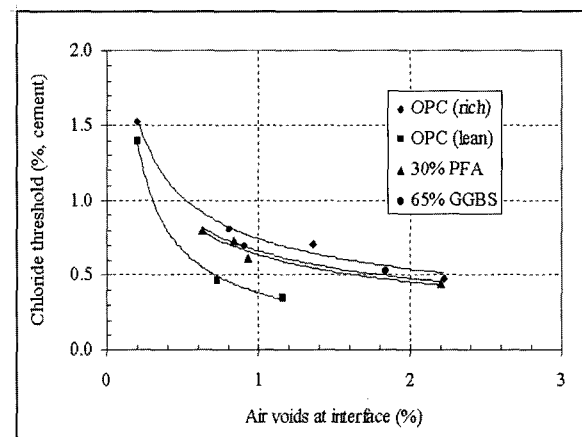


Fig. 5 Relationship between the air void content at the steel-concrete interface and the CTL expressed as a percentage of the cement weight

content, around 0.2% in this study.

The CTL for concrete containing 30% PFA and 65% GGBS ranged from 0.44 to 0.8% by weight of cement, corresponding to air void volumes ranging from 0.63 to 2.2%. At a given air void content, the CTL for concrete containing 30% PFA or 65% GGBS was slightly lower than those for an OPC rich mix and higher than an OPC lean mix. The results showed that PFA and GGBS additions had little beneficial effect in raising the CTL.

3.2 Transport of chloride

The apparent diffusion coefficient and the concentration of chloride ion at the concrete surface were calculated to assess the transport of chloride in concrete. The plot of diffusion coefficient and surface concentration as a function of binder type and interfacial air void ratio is shown in Figs. 6 and 7.

The apparent diffusion coefficients for OPC, 30% PFA and 65% GGBS concrete are shown in Fig. 6. For each mix, the diffusion coefficient increased as the interfacial voids increased, except for 30% PFA concrete where the

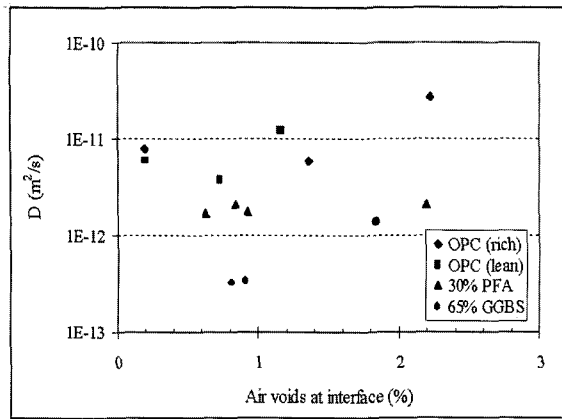


Fig. 6 Diffusion coefficients for OPC, 30% PFA and 65% GGBS concrete as a function of void content at the steel-concrete interface

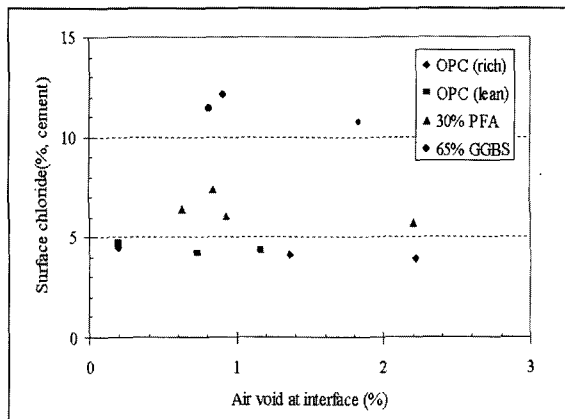


Fig. 7 Surface chloride concentration of OPC, 30% PFA and 65% GGBS concrete as a function of air void content at the steel-concrete interface

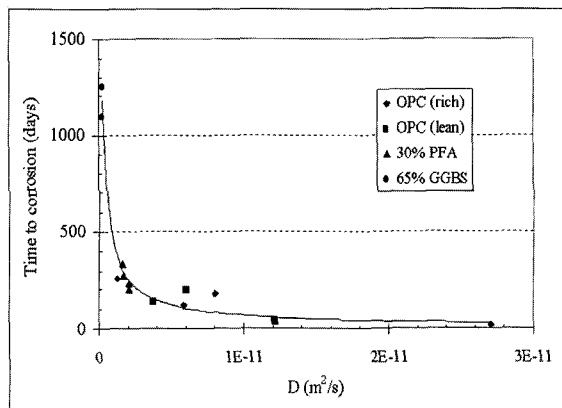


Fig. 8 Relationship between the diffusion coefficient and time to corrosion initiation

coefficient was relatively constant. For OPC concrete, the apparent diffusion coefficient for a rich mix ranged from 8.05×10^{-12} to 2.71×10^{-11} m^2/s , which was slightly higher than for a lean mix with values ranging from 5.99×10^{-12} to 1.22×10^{-11} m^2/s . A 65% GGBS concrete showed the lowest diffusion coefficient, ranging from 1.36×10^{-12} to 3.36×10^{-13} m^2/s . It seems likely that poor compaction in the casting of the concrete resulted in a more open pore structure which favoured the transport of chloride ions.

The surface chloride contents measured are plotted in Fig. 7. The surface chloride varied with the cementitious binder, whereas the interfacial void content appeared to have no effect on to the surface chloride content. 65% GGBS and 30% PFA concrete had a higher surface chloride concentration than OPC. For OPC, the surface chloride ranged from 3.91 to 4.74% of the cement weight, while 65% GGBS concrete showed values from 10.71 to 12.07%. However, the quantities of chloride ions for each mix are similar, in the range 12 to 18 kg/m^3 , and the surface chloride is therefore likely to be influenced more by the duration of exposure to the chloride environment rather than to binder type.

3.3 Time to corrosion initiation

The time to corrosion initiation is dependent on the rate of chloride ingress, which can be determined from the diffusion coefficient and the CTL. Thus, the higher the chloride diffusion coefficient, the shorter the time to corrosion initiation. Fig. 8 shows that for all specimens, an increase in the diffusion coefficient produced a reduction in the time to corrosion initiation. Also, at a given diffusion rate, the time to corrosion was strongly dependent on the CTL. It was found that the time taken for the passive film to breakdown varied from 20 to 1,250 days, depending on the cementitious binder type and the air void content at the steel-concrete interface. For OPC, the time to corrosion initiation ranged from 20 to 199 days, which was the lowest level found in this study. It was also observed that corrosion initiation started slightly earlier in rich compared to lean mix concretes, which may be due to the difference in chloride transport conditions in concrete. It is well known that the diffusion of chloride is more rapid in cement-rich concrete.⁹⁾

Replacement with 65% GGBS resulted in a significant increase in the time to corrosion initiation, in spite of the small increase in the CTL. The time to corrosion initiation varied from 255 to 1,250 for 65% GGBS concrete, compared to 200 to 331 days for 30% PFA concrete and less than 200 days for OPC concrete. This effect is attributed to the resistance to the transport of chloride ions in concrete (lower diffusion coefficients).

4. Discussion

4.1 Influence of binder type

The cementitious binders investigated in this study have a much smaller impact on the CTL than the entrapped air void content at the steel-concrete interface. 30% PFA and 65% GGBS concretes have a similar or lower range CTL compared to OPC.

The effect of mineral admixtures such as PFA and GGBS replacement on the CTL has been the subject of many

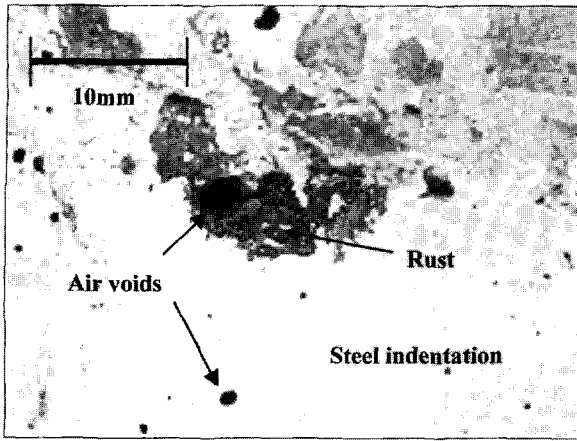


Fig. 9 Steel-concrete interface showing an early stage of corrosion

debates in the literature. The binding capacity of GGBS or PFA is higher than OPC and hence a higher CTL is expected¹⁰⁾. However, other research has shown that PFA concrete lowered the CTL¹¹⁾. In the present study, the higher binding capacity of GGBS and PFA did not result in higher or lower values of the CTL. The results suggest that the CTL may not be affected by the chloride binding of mineral admixtures, but by one of the physical properties of concrete, such as the air void content at the steel-concrete interface. However, the remarkable effect of PFA and GGBS in enhancing the resistance to chloride transport is notable. In particular, 65% GGBS concrete produced very dense concrete, in which the diffusion coefficient of chloride was lower when compared to OPC by 2 orders of magnitude. A significant decrease in the diffusivity of GGBS concrete resulted in a much longer time to corrosion initiation in spite of the lower CTL. For example, a 65% GGBS concrete in this study showed a time to corrosion initiation of 1,250 days with 0.8% of the CTL, compared to 130 days for OPC with the same CTL level. It is concluded that the beneficial effect of mineral admixtures cannot be attributed to the higher CTL, but rather to a lower chloride transport.

The present study has shown that an increase in cement content resulted in a higher CTL. A higher cement content and a lower water-cement ratio have been generally advised to prevent the corrosion of steel because the cement hydration products inhibit corrosion by buffering a pH fall^{12,13)}. However, an increase in the cement content may offset its beneficial effect in increasing the CTL. In this study, a reduction in cement content at a constant water-cement ratio, resulted in an increase in the resistance to the transport of chloride. It was observed that for a lean mix, the time to corrosion initiation was slightly longer when compared to a rich mix, although the CTL of the lean mix was lower than that of the rich mix. With regard to water absorption, a lower rather than higher cement content is again advantageous. A reduction in cement content, at a given water-cement ratio

will reduce the amount of water absorption and hence chloride ingress if the water contains chloride ions. This suggests that optimising the minimum cement content as a function of the CTL and the chloride transport is required in order to decrease the chloride transport and thus to increase the corrosion-free life at a given CTL.

4.2 Air voids at the steel-concrete interface

The importance of entrapped air voids adjacent to the embedded steel has been emphasised, because corrosion starts there. It has been observed that air voids are often generated by bleeding or/and settlement underneath the embedded steel, perpendicular to the direction of casting, and then corrosion initiates in these voids irrespective of whether chlorides are introduced internally or externally.¹⁴⁻¹⁶⁾ It is due to the fact that voids, in the vicinity of the steel, saturated in the pore solution would provide the more active environment for electrochemical reaction (i.e. corrosion process) than cement matrix that relatively restricts the flowing current between anode and cathode, because of higher electrical resistance.

In replicate specimens, poor compaction and vibration in the casting of concrete specimens was found to increase the air void content. Entrapped air voids do not influence chloride diffusion because they do not build up a network to allow the transport of solution ions. However, the entrapped air voids at the steel-concrete interface do not contain cement hydration products that would otherwise chemically bind or physically adsorb chloride ions. If the void is dry, its influence on corrosion of the reinforcing steel will be marginal. A void at the interface which is saturated with pore water will provide anodic sites where corrosion will be initiated. Fig. 9 shows a steel-concrete interface after corrosion initiation.

The presence of a rust stain at an entrapped void would appear to support the hypothesis that corrosion initiation takes place at the void-steel interface, and in turn would influence the CTL.

The time to corrosion may also be influenced by the air void content at the steel-concrete interface. However, this effect is not only because the void itself affects the chloride transport, but also because the CTL is strongly influenced by the air void content. It seems that the void content influences the transport of chloride ions in concrete. Diffusion coefficients for all the specimens were increased by an increase in the void content, whereas the change in surface chloride content with the void content is negligible. This implies that the entrapped air voids may be largely saturated with pore water and thus they might be expected to accelerate the diffusion of chloride ions. This study on the interfacial void suggests that reducing the entrapped air void

5. Conclusions

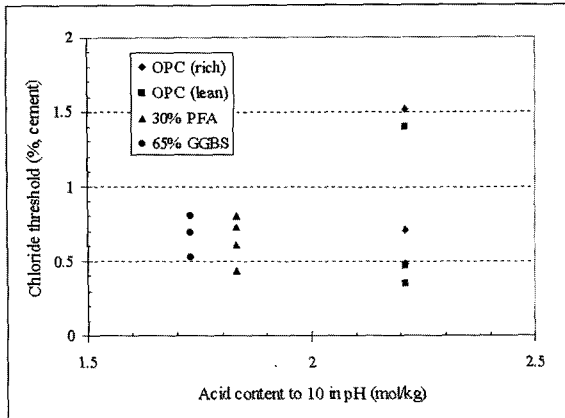


Fig. 10 Relationship between the ANC at pH 10 and CTL for OPC, 30% PFA and 65% GGBS concretes.

content will increase the CTL, decrease the diffusion of aggressive ions from an external source, and thus significantly prolong the corrosion-free life of concrete structure.

4.3 Buffering capacity

The passive film breakdown is accompanied by acidification at the anodic site and thus most bound chlorides are released by a fall in pH at the stage of corrosion initiation.

This supports the use of the total chloride content to represent CTL, and also suggests that the rate at which bound chloride is released may be more important than binding capacity when considering the CTL. The rate of release of bound chloride can be expressed as an acid neutralisation capacity (ANC) of cement matrix, which was introduced to concrete technology by Glass and Buenfield.¹⁷⁾ In this study, the ANC of each binder was not measured but was obtained from the data of Reddy.¹⁸⁾

Fig. 10 gives the relationship between the ANC of OPC, 30% PFA and 65% GGBS concrete and the CTL. The ANC at pH 10, at which 98% of chlorides are generally free, was 2.21, 1.83 and 1.73 mol/kg for OPC, 30% PFA and 65% GGBS concrete specimens respectively. A pH below 10 is necessary to produce a thermodynamically unstable passive film. Although results in the literature indicate a much higher chloride binding capacity for PFA or GGBS concrete, their ANC level is lower than that of OPC. An increase in the ANC level also resulted in an increase in the CTL. The variance in the ANC results for replicates seems to be due to the interfacial air void ratio at the steel-concrete interface, and suggests that the lower buffering capacity of mineral admixtures may present a higher corrosion risk even with a smaller chloride content.

- 1) The CTL measured in this study for OPC, 30% PFA and 65% GGBS concretes was found to be in the range 0.35 to 1.52, 0.44 to 0.8 and 0.53 to 0.8% by weight of cement respectively. The effect of the mineral admixture on the CTL was found to be relatively small.
- 2) The entrapped air voids at the steel-concrete interface resulting from different levels of compaction significantly influenced the CTL: an increase in the air void content resulted in a decrease in the CTL regardless of cementitious binder type. In particular, the CTL increased remarkably when the air void content decreased to below 0.5%.
- 3) With regard to the cement content, a higher cement level for a given water-cement ratio (0.45 in this study), resulted in an increase in the CTL and diffusion coefficient of chloride. This combination of higher CTL and diffusion coefficient reduced the time to corrosion initiation. The CTL was found to be similar for both rich and lean concrete mixes at very low void contents.
- 4) For the same mix, the time to corrosion initiation is also influenced by the air void content and the CTL. However, the more crucial factor is the diffusion coefficient, which varied with binder type. The order of the time to corrosion initiation was found to be 65% GGBS > 30% PFA > OPC (lean mix) > OPC (rich mix).
- 5) The effect of binding capacity on the CTL was found to be marginal. In spite of the high binding capacity level in the 65% GGBS concrete, its CTL was slightly lower than that of OPC (rich mix). The ANC at pH 10 was found to have more influence on the CTL than the binding capacity.

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