

The influence of blast furnace slag, fly ash and silica fume on corrosion of reinforced concrete in marine environment

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Chloride penetration from sea water may cause corrosion of reinforcement in concrete structures. Adding reactive inorganic materials such as blast furnace slag, fly ash or silica fume to the cement matrix improves the resistance against chloride penetration as compared to Portland cement concrete. A relatively simple laboratory procedure was proposed to test concrete mixes for their expected service life in sea environment. The assumption is that chloride diffusion and cover depth determine the time-to-initiation of corrosion and electrical resistivity determines the corrosion rate and so the time-to-cracking. The sum is the total "service life".

The proposed testing procedure was applied to five concrete mixes. They contained ordinary Portland cement; OPC + silica fume; OPC + fly ash + silica fume; blast furnace slag cement, all with gravel aggregate; OPC and lightweight coarse aggregate. The predicted time-to-initiation and the time-to-cracking in marine splash zone were calculated from the test results.

The calculated service lives range from 10 to 80 years, increasing in the order: OPC-lightweight aggregate < OPC-gravel < OPC + silica fume-gravel < OPC + fly ash + silica fume-gravel < blast furnace slag cement-gravel. The ranking and the numerical results are in general agreement with data from exposure research and with practical experience. The addition of blast furnace slag, fly ash and/or silica fume may significantly increase the resistance against chloride penetration and consequently the time-to-initiation of corrosion. Once corrosion has started, steel in concrete with additional cementing materials may have a decreased rate of corrosion.

The proposed set of laboratory tests appears suitable for evaluating the effect on corrosion protection in marine environment of new and/or supposedly improved concrete compositions.

Keywords: concrete, reinforcement corrosion, marine environment, diffusion, resistivity, service life, durability

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Introduction

Penetration of chloride from sea water is the main cause of depassivation of steel reinforcement and subsequent damage to marine concrete structures, in particular in the splash zone. The usual precautions are a low water-to-cement ratio (w/c) and a thick concrete cover to the steel. These have practical limitations, however. Carbonation induced corrosion is usually prevented by a low w/c and sufficient cover. World-wide, many structures in chloride containing environment with low w/c and thick cover suffer from early corrosion damage. In marine environment, the penetration of chloride is the most important problem. Obviously, in all cases adequate cover thickness and compacting of the fresh mix are essential.

A high chloride penetration resistance may be obtained by adding hydraulic or pozzolanic minerals to the cement matrix. Examples are blast furnace slag (BFS or ground granulated blast furnace slag, GGBS) which has been used in concrete in The Netherlands for over 60 years (Wiebenga 1980), and silica fume (SF, microsilica) and fly ash (pulverised fuel ash, PFA) which are being used in other countries. These materials are called additional cementing materials and the resulting binders are called blended or composite cements. The effect of the additions is that the pore structure becomes finer, most importantly of the bulk paste (Larbi 1993); possibly the chloride binding capacity is higher. Consequently, the main positive effects of these additions are (COST 1996):

- slower chloride penetration by 3 to 10 times as compared to Portland cement concrete;
- similar or higher critical chloride content (which starts corrosion);
- similar or lower corrosion rate (once corrosion has started).

These positive effects have been demonstrated by numerous field and laboratory studies. Some examples will be given in a following section.

Possible negative effects are:

- lower strength at young age;
- more sensitive to poor curing;
- more critical mix design for workability.

The negative effects can be minimized by proper mix design, making mix trials and applying quality control of raw materials and concrete on site. Further developments of composite cements are in progress (like rapid hardening blast furnace slag cement).

Although a large amount of positive experience exists on the use of blended cements, it is not easy to evaluate. There are only a limited number of field surveys of concrete made with blended cements. Wiebenga (1980) describes 50 years of experience with slag cement in coastal construction in The Netherlands. The amount of corrosion damage in blast furnace slag cement structures was low (one case with much corrosion, one with moderate corrosion, both after 45 years, out of 48 inspected structures with slag cement). There is relatively little comparative testing under practical conditions. Polder & Larbi (1995, 1996) describe tests of OPC and BFS concrete after submersion in the North Sea for 16 years. They have shown that chloride penetration is much less in slag cement concrete than in OPC concrete. In a recent monograph, Bijen has reviewed the experience with slag cement concrete (Bijen 1996). In addition to reinforcement corrosion, other durability aspects are treated, such as resistance against carbonation, sea water and salt attack, alkali-silica reaction and

freeze-thaw (deicing salt) action. The durability of the slag cement used in the King Fahd Causeway, Saudi Arabia is described in detail. Overall, the durability of slag cement concrete is shown to be excellent.

Bamforth and co-workers have published on splash zone exposure of various concrete mixes since 1987 (Bamforth & Chapman-Andrews 1994), including fly ash, slag and silica fume. The results have shown that chloride penetration is less in blended cement concrete as compared to various OPC concretes. The British Building Research Establishment has exposed concrete with various fly ash contents to marine environment. Thomas (1996) has reported that the critical chloride content decreases with higher fly ash contents in particular over 30%. For fly ash dosages less than or equal to 30%, corrosion initiates later. From these finding and despite the possibly lower chloride threshold, he states that fly ash concrete provides better protection.

However, one field test or case study is difficult to compare to another. For the designing engineer, a quick and simple but still realistic test would be useful, for instance to compare alternative mixes for a particular structure. A set of laboratory tests was proposed for evaluating the durability of the corrosion protection in marine splash zone environment of a specific (blended cement) mix in a test period up to two years (Polder 1990). Five concrete mixes were tested accordingly in a study commissioned by SMOZ and CUR. For more details see (Polder 1996). This paper describes the model behind the tests and the results of the five mixes.

Reinforcement corrosion in marine environment

According to Bažant (1979) and Tuutti (1982), the service life is the sum of the corrosion initiation period and the corrosion propagation period. In a marine environment, the initiation period is determined by chloride penetration through the concrete cover to the reinforcement, until the steel loses its passivation and corrosion starts. The propagation period is characterised by the rate of corrosion and the amount of corrosion that causes the concrete cover to crack. Major cracking of the concrete cover is considered to indicate the end of the service life; generally, maintenance action will be taken then. Without maintenance, structural safety may become impaired in a relatively short length of time. Some authors do not include the propagation period in the design service life. In cases where the corrosion rate is high, the difference is negligible. In cases with a relatively low corrosion rate, the length of the propagation period may be significant. Including the propagation period in the design life in such cases has economic benefits.

SMOZ: Foundation for Materials Research in the Sea, Bilthoven, The Netherlands
CUR: Centre for Civil Engineering Research and Codes, Gouda, The Netherlands

Within a marine structure, the risk of corrosion is generally largest in the splash zone, because both chloride penetration and corrosion rate are relatively rapid. Chloride penetration in the surface concrete may be accelerated by partial drying out of the concrete and subsequent capillary absorption of sea water. The corrosion rate after depassivation is comparatively high because the concrete remains quite wet due to the frequent exposure to splash, so the concrete resistivity is relatively low. Furthermore, because the concrete is not completely water saturated, sufficient oxygen can reach the steel. In permanently submerged concrete, chloride penetration may be quick, but corrosion does not occur due to lack of oxygen. The tidal zone is either similar to the submerged zone or to the splash zone, depending mainly on how quick the concrete dries out during the dry part of the tidal cycle. In the atmospheric zone, the chloride load will be less than in the splash zone, so penetration to the reinforcement will take much longer.

Laboratory testing for corrosion protection

According to the procedure described in this paper, the expected durability of the corrosion protection is calculated from the chloride diffusion plus cover depth (resulting in the time-to-depassivation) and the electrical resistivity plus tensile strength (resulting in the time-to-cracking) of each mix, using a modification of the life time model described by Bažant (1979). The calculation will be explained in the following sections.

Corrosion initiation

In the test procedure, it is assumed that the transport of chloride through concrete may be modelled as a diffusion process, which is justified for the following reasons. Firstly, the effect of capillary absorption is only significant at low cover depths and on the short term. In the longer term and at higher cover depths, chloride transport will be dominated by diffusion. This was confirmed from concrete exposure to marine splash for 6 years (Bamforth & Chapman-Andrews 1994), where the penetration was modelled as diffusion and coefficients were obtained which are similar (for similar concrete mixes) to those from 16 years submersion in the North Sea (Polder & Larbi 1996). Secondly, it was shown experimentally that the influence of chloride binding by cement components may be included in the effective diffusion coefficient (Sergi et al. 1992, Yu et al. 1993).

For the input values of the calculation the following data were regarded. The surface chloride concentration C_s was estimated by Sørensen (1979) from measurements on marine structures to be about 5% chloride by mass of cement. Based on splash zone exposure tests, Bamforth & Price (1993) suggest to use a value of 4.5% for service life design calculations. From the present laboratory tests slightly lower C_s values were found, namely between 3 and 4%. Consequently, 4% is taken as the value for C_s in the calculations.

Regarding the critical chloride content (C_{cr} , or threshold value), values about 0.4% by mass of cement have been adopted in many national standards as acceptable for new concrete. Recently it has been established that significant corrosion rates in alkaline (i.e. non-carbonated) concrete do not occur at chloride contents less than 1% (Bakker et al. 1994). It appears that 1.0% chloride by mass of

cement is a reasonable, possibly even conservative, value for good quality concrete such as used for most marine structures (and for the mixes investigated here), provided concrete cover to the steel is adequate.

If D_{eff} of a particular concrete mix is known from experiments and using Fick's second law of diffusion with the specified values for C_s and C_{cr} , for a given cover depth L the time-to-initiation t_i can be calculated with:

$$t_i = L^2 / (2.65 \cdot D_{eff}) \quad (1)$$

with L in m, D_{eff} in m^2/s , giving t_i in seconds.

In the proposed test set up, the effective chloride diffusion coefficients D_{eff} is calculated from diffusion profiles that are fitted to chloride profiles that are determined after 1.5 years of submersion in 3.5% NaCl solution at room temperature and slightly elevated temperatures.

Corrosion propagation

After depassivation, the corrosion rate is assumed to be determined by the resistivity of the concrete. This is a simplification, for which there is experimental and theoretical support. It may be assumed that the anodic process is not rate-controlling at chloride contents over 1% (Bakker et al. 1994). The cathodic process is not rate-controlling because the concrete is not completely water saturated, which allows sufficient oxygen to penetrate to the steel. From macrocell experiments, Raupach (1992) found that the resistivity is the major factor controlling the corrosion rate for concretes with a relatively high resistivity. In low resistivity concretes, other factors appeared to be dominating the corrosion rate. Pitting corrosion may be regarded a special case of macrocell corrosion, with anode-to-cathode distance smaller than in a true macrocell. A linear relation between the inverse of concrete resistivity and steel corrosion rate has been established from steel polarisation resistance experiments in mortars involving various cement types in the laboratory (Alonso et al. 1988, Lopez & Gonzalez 1993, Glass et al. 1991). From experiments on concrete blocks exposed to marine splash for 6 years, a similar relationship between resistivity and corrosion rate was found (Polder et al. 1994), suggesting resistive control for resistivities greater than about 70 $\Omega.m$. Using these data, Bazant's theoretical model was calibrated. His original equations appeared to produce very high corrosion rates, probably due to an overestimated maximum potential difference between anodic and cathodic sites. Using the calibrated model, the corrosion rate CR is given by:

$$CR = \frac{1000}{\rho_{concrete}} \quad (2)$$

with $\rho_{concrete}$ the concrete resistivity in $\Omega.m$, giving CR in $\mu m Fe/year$.

To calculate the length of the propagation period t_p from a calculated corrosion rate, the amount of corrosion that causes cracking must be known. From practical observations, Tuutti (1982) found

that the critical amount of corrosion for bars at 10 mm cover depth was 200 μm . Experiments and calculations by Andrade et al. (1993) have shown that very fine cracks are caused by as little as 20 μm corrosion, and cracks wider than 0.3 mm originate from 100 to 200 μm of steel thickness loss. Considering the stresses caused by the expansion and the elasticity of concrete (taking into account creep), Bazant (1979) has stated that the critical amount of corrosion is proportional to the tensile strength of the concrete. His calculated results are in general agreement with the other data. The present approach arrives at values for steel loss that cause cracking between 45 and 65 μm . It is recognised, however, that there is some debate on the amount of steel loss that causes cracking, for example as a function of concrete quality, moisture content, cover depth and cause of corrosion.

The length of the propagation period t_p can be calculated from the amount of corrosion that causes cracking divided by the corrosion rate. Assuming that the corrosion rate is determined by the resistivity according to (2), this results in:

$$t_p = 12.5 \cdot 10^{-12} \cdot f_t \cdot \rho_{\text{concrete}} \quad (3)$$

with t_p in year and f_t the tensile strength of the concrete in N/mm^2 .

In the proposed test set up, the resistivity is determined as follows. Four small prisms with embedded measuring electrodes are exposed in a fog room and the resistivity is determined using alternating current. The tensile strength is determined as splitting tensile strength of standard cubes after 28 days of curing in a fog room.

Service life calculation

The service life of each mix is then calculated by adding the initiation period to the propagation period:

$$t_i = t_i + t_p \quad (4)$$

Experimental programme

Concrete mixes investigated

The five investigated concrete mixes included three mixes with additional cementing materials and one plain Portland cement, all with river sand and gravel, one with lightweight (sintered fly ash) aggregate and river sand mix. All mixes had a low w/c of 0.43, such as specified by the Dutch concrete technology standard (VBT 1986) for marine environments. The main compositional features and the mix codes are given in Table 1. A superplasticiser was added in such a dosage that a slump of about 100 mm was obtained. Such a slump would be necessary in practice to allow sufficient compacting in locations with high rebar densities. For the Lytag mix, the slump was kept slightly lower in order to prevent floating of lightweight aggregate particles. The Lytag was prewetted before mixing.

Table 1. Concrete compositions and fresh concrete slump.

concrete type	mix S	mix L	mix F	mix P	mix B
cement type	OPC	OPC	OPC	OPC	BFSC (Portland blast furnace cement CEM III/B 42.5)
specific features	5% silica fume	-	5% silica fume & 10% fly ash	-	70% slag
coarse aggregate	siliceous, river gravel D 16 mm	Lytag D 12 mm	siliceous, river gravel D 16 mm	same	same
sand	all	siliceous	river	sand	
cement content (kg/m ³)	340	353	337	339	338
water content (kg/m ³)	146	152 ¹	145	146	145
water-to-cement ratio	0.43	0.43 ¹	0.43	0.43	0.43
superplasticiser (% by cement mass)	2.65	2.65	3.06	2.47	2.02
slump (mm)	100	80	100	120	125

¹ exclusive of 10% (m/m) absorbed water with respect to Lytag mass

Test methods

For each mix, compressive and tensile strength of three 150 mm cubes and density of six cubes were determined after 28 days storage in a fog room according to the relevant Dutch concrete testing standards.

One prism of 100 x 100 x 300 mm³ for each mix was submerged in 3.5% NaCl solutions at 30 and 40°C, respectively, and one prism of the OPC mix also at 20°C. After 1.5 year submersion, thin layers of a few millimetres were ground (dry) from the prisms. The powder was analysed for total chloride content by dissolution in concentrated hot nitric acid and chloride determination by Volhard titration. Diffusion curves were fitted to the chloride profiles and chloride diffusion coefficients were calculated.

The electrical resistivity of the concrete was determined on specimens of 100 x 100 x 50 mm³ prisms with two embedded electrodes at 50 mm spacing. Four prisms for each mix were exposed during

1.5 year in a fog room and four in air at 20°C and 80% relative humidity, respectively. After that time the two sets of specimens were exchanged between the two climates and measurements were carried on until 2.4 years. The electrical resistance between the two electrodes was measured using 108 Hz Alternating Current. The resistivity (specific resistance) was calculated by multiplying with a cell constant (0.063 m) which was obtained from calibration of a cell with identical geometry and electrolyte solutions of known conductivity.

Results and discussion

The results of the mechanical testing and the densities at 28 days are given in Table 2. The mixes S and F have higher strengths (by about 30%) than mix P, apparently due to the addition of silica fume. Regarding strength, mix B is completely equivalent to mix P. Mix L has a normal compressive strength but a slightly lower tensile strength and expectedly, a lower density. All other mixes had similar densities.

Table 2. Mechanical properties and densities of concrete after 28 days curing in a fog room.

concrete type	mix S	mix L	mix F	mix P	mix B
compressive cube strength (N/mm ²)	64.1	49.1	68.2	50.4	51.2
% of mix P	127	97	135	100	102
splitting tensile strength (N/mm ²)	5.2	3.5	4.9	3.9	3.9
% of mix P	134	91	127	100	99
density (kg/m ³)	2412	2037	2413	2431	2415
% of mix P	99	84	99	100	99

The fitted chloride penetration profiles after 1.5 years of exposure to salt solution of 30°C are given in Figure 1. The calculated effective diffusion coefficients for all temperatures are presented in Table 3. The effect of increased temperature is practically absent for individual mixes. The differences between the five mixes are large. The OPC mixes P and L (made with gravel or Lytag, respectively) have a relatively high chloride diffusion coefficient. Of these, Lytag concrete has a higher diffusivity than gravel/OPC concrete. On the other hand, the composite (blended) cement mixes B (blast furnace slag) and F (silica fume and fly ash) have a diffusion coefficient which is about three times lower than for mix P. Only the silica fume mix S, which is a blended mix, does not fit into this pattern. It was expected to have low chloride diffusion, but it is similar to the plain OPC mix. Possibly a higher level of addition would have resulted in increased chloride penetration resistance. It is clear from the increased strength (compared to mix P) that the silica fume has had some effect on the microstructure of the concrete.

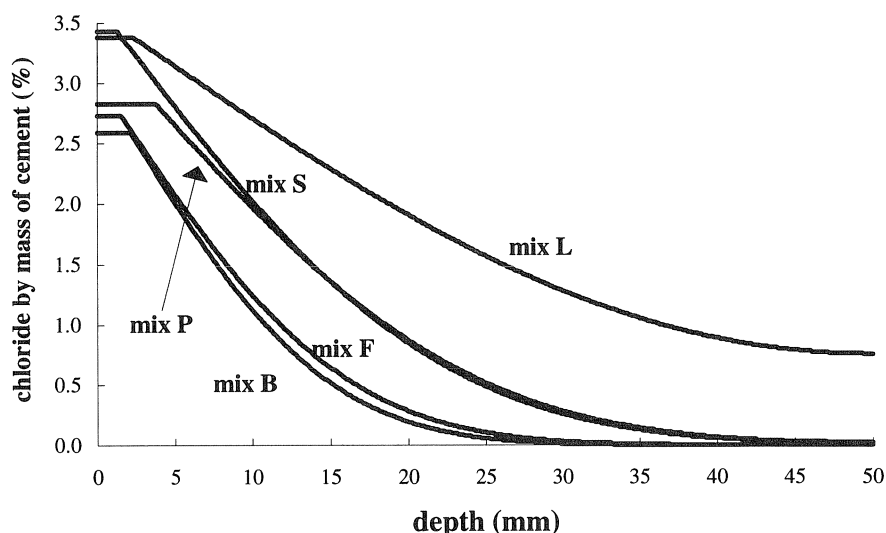


Fig. 1. Fitted chloride diffusion profiles for the five mixes after 1.5 year submersion in 3.5% NaCl solution at 30°C.

The diffusion coefficient for mix P is quite similar to values found after 16 years submersion in the North Sea for OPC concrete (Polder & Larbi 1996). Blast furnace slag concrete diffusion coefficients after 16 years were about 2.5 times lower than the present mix B values (0.3 versus $0.8 \cdot 10^{-12}$). This is probably caused by further hydration of the slag during the long wet exposure (Bamforth 1994). Considering this, both mix P and mix B results are in general agreement with data from long term natural sub-sea exposure and may be regarded as reliable values for marine (diffusion) conditions. Apparently the lower temperature in the sea exposure (approx. 10°C) than in the laboratory (20 to 40°C) has no strong influence on the diffusion coefficient.

A remark must be made regarding the observed diffusion in mix L. The Lytag was wetted before mixing the concrete. During the tests, the material remained water saturated. The diffusion in mix L concrete is about three times faster than in mix P. As mix L and mix P have the same matrix, the higher diffusivity is probably due to transport through (water filled) channels in the porous aggregate. In real splash zone conditions, the concrete and in particular the pores in the aggregate particles would dry out to a certain extent. Once the rather coarse channels have dried out, they may not become (completely) saturated again and the transport could not proceed through the aggregate anymore. Consequently, the effective diffusion in practical splash zone conditions of mix L and porous aggregate concrete in more general, would be slower than indicated by our tests, probably approaching the value of mix P.

Table 3. Effective chloride diffusion coefficients after 1.5 year exposure to 3.5% NaCl solution for various exposure temperatures (20°C only mix P) and overall result for each mix.

concrete type	mix S	mix L	mix F	mix P	mix B
temperature	diffusion coefficient (* 10 ⁻¹² m ² /s)				
40°C	3.2	7.7	1.0	2.5	0.7
30°C	2.4	8.4	1.2	2.5	1.0
20°C	-	-	-	2.5	-
overall	3	8	1.1	2.5	0.8

- not tested

The resistivities as a function of exposure time are shown in Figure 2 (for specimens that started in the fog room) and values after 1.5 year are given in Table 4. Most results were fairly stable between 3 months and 1.5 year. The resistivities in 80% RH were much higher than the “wet” values. Upon transfer of the “dry” samples to the fog room after about 1.5 year, the resistivities decreased roughly to their “wet” values. Only the value of mix L was significantly higher when transferred to the fog room after exposure in 80% RH as compared to constantly wet specimens (+35%). This is probably due to drying out of the pores in the aggregate which are apparently not fully rewetted. Because in the splash zone at least partial drying out will occur, both values for the resistivity are used for the service life calculation.

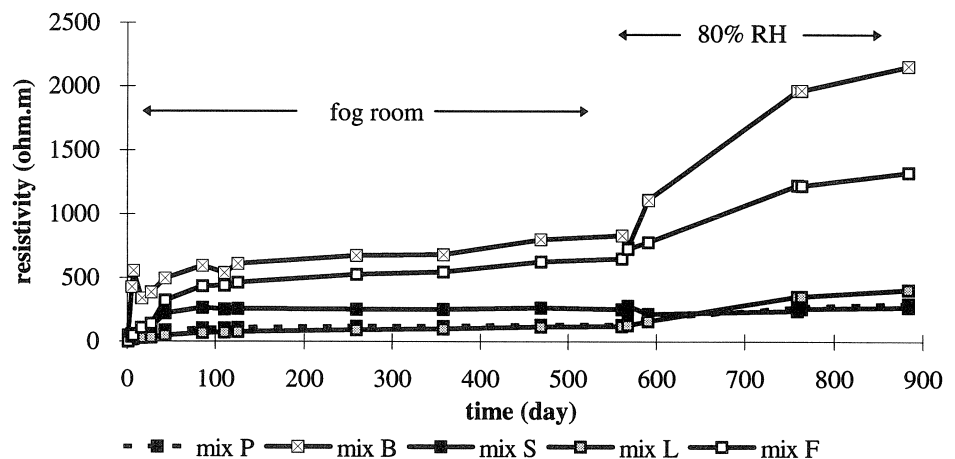


Fig. 2. Concrete resistivity for five mixes during exposure in a fog room for 560 days and another 320 days in air of 20°C and 80% RH.

For all concrete mixes in the fog room, the resistivity was greater than 100 $\Omega\cdot\text{m}$ from an age of one year on. This fulfils the condition that resistivity must be greater than 70 $\Omega\cdot\text{m}$, which allows calculating the corrosion rate from the resistivity. The observed resistivities correspond well to laboratory (Polder & Ketelaars 1991) and field (Polder et al. 1994) values for concrete mixes of similar composition (cement type, water-to-cement ratio).

Service life calculation

The properties found for the five investigated mixes in the various experiments were used for the service life calculation. One more datum is needed, namely the cover depth. As a normal value for marine construction, 50 mm is chosen. The data and the results in terms of duration of initiation and propagation period are presented in Table 4. As mentioned before, for mix L two resistivities ("wet" and "dry \rightarrow wet") are given and two service life values are calculated. The difference in terms of propagation period is small, however.

Table 4. Service life calculation results for a cover depth of 50 mm.

Concrete type	mix S	mix L	mix F	mix P	mix B
D_{eff} ($\cdot 10^{-12} \text{ m}^2/\text{s}$)	3	8	1.1	2.5	0.8
t_i (y)	10	3.7	30	12	37
f_t (N/mm^2)	5.2	3.5	4.9	3.9	3.9
ρ_{concrete} ($\Omega\cdot\text{m}$)	250	120–160	650	135	830
t_p (y)	16	5–7	40	7	40
t_1 (y)	26	9–11	70	19	77

Conclusions

Investigated concrete mixes

According to the described tests and calculations, concrete mixes made with blended (composite) cements containing 70% blast furnace slag (mix B) or 10% fly ash plus 5% silica fume (mix F) have improved durability properties in marine environment. Service lives can be obtained that are several times longer than with plain Portland cement concrete. According to the test adding only 5% silica fume (mix S) to Portland cement does not improve the durability considerably. Using porous lightweight aggregate (and Portland cement, mix L) appears to cause earlier depassivation of the reinforcement than using dense gravel aggregate. This may be an underestimate due to the simplifications in the test method.

The calculated service lives in the splash zone range from about 10 to about 80 years, with increasing service life in this order: lightweight aggregate/OPC (mix L) < gravel/OPC (mix P) < gravel/silica fume/OPC (mix S) < gravel/silica fume/ fly ash/OPC (mix F) < gravel/blast furnace slag (mix B). This ranking is in general agreement with expectations from concrete exposure research and with practical experience. Quantitatively, the service life results are in the range that is observed in practice (as far as experience exists), so the prediction is reasonable.

Other blended cement compositions

From various studies it is clear that using fly ash alone (without silica fume) as an additive can have similar effects as the blended mixes tested here. The dosage should be about 25 to 30% fly ash to total cementitious mass in order to obtain a similar improvement of the expected durability as with 70% slag or with 10% fly ash and 5% silica fume.

The dosage of silica fume is critical in the respect that too low dosage does not cause the desired improved durability (such as in mix S in this work), but too high dosage may have negative side effects such as lowering the pH of the pore solution, and so lowering the critical chloride content. Possibly 8% silica fume is about optimal. Such a narrow “good” composition range makes proper mixing of silica fume into fresh concrete rather critical. In particular under field conditions, this may be difficult. The present results have shown that a higher strength (such as obtained with 5% silica fume), does not automatically mean that the durability is improved.

Laboratory testing for corrosion protection

The service life calculation based on chloride diffusion rate plus cover depth (time-to-depassivation) and concrete resistivity plus tensile strength (time-to-cracking) was concluded to produce useful and realistic durability predictions for concrete in marine splash zone. The simple proposed laboratory experiments with a duration of between one and two years appear to produce suitable results for this calculation.

Chloride penetration was determined satisfactorily from constant submersion tests and subsequent determination of the chloride profile by dry grinding and analysing thin layers. A suitable exposure temperature may be 20 or 30°C. The effect of higher temperature is negligible.

Measuring the electrical resistivity of the concrete in wet conditions between a half and two years age gives useful information for estimating the corrosion rate.

Because the model used for chloride penetration is more firmly based and verified in the field than the model used for the corrosion rate, the calculated time-to-initiation results are probably more accurate than the time-to-cracking results.

The proposed test set up is suitable for testing new and/or supposedly improved concrete compositions for their effect on corrosion protection in marine environment.

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