Study of chloride penetration profiles and surface chloride content of concrete structures exposed to marine environment evaluated at different exposure times

Cristiane Silva (Main Author)
Universidad Federal de Rio Grande, Departamento de Ingenieiría Civil
Rio Grande, 96500 (Brazil)
crisarpino@ibest.com.br

Bernal Camacho Jesus (Corresponding Author)
Universidad Autónoma de Sinaloa, Escuela de Ingeniería Mazatlán
Sinaloa, 82017 (Mexico)
jmbernalc@uas.edu.mx

Júlio Bandeira
Universidad Federal de Rio Grande, Departamento de Ingeniería Civil
Rio Grande 96500 (Brazil)
jorge-band@hotmail.com

André Guimarães
Universidad Federal de Rio Grande, Departamento de Ingenieiría Civil
Rio Grande, 96500 (Brazil)
atcg@vetorial.net

Ramón Corral Higuera
Universidad Autónoma de Sinaloa, Facultad de Ingenieria Mochis
Sinaloa, 81223 (Mexico)
ramon.corral@uas.edu.mx

Manuscript Code: 1461
Date of Acceptance/Reception: 02.12.2019/03.05.2019
DOI: 10.7764/RDLC.18.3.545

Abstract

Nowadays, some studies have shown the increasing chloride content of concrete surface (Cs) exposed to marine environments over time. To evaluate this behavior, a comparative analysis was conducted between the chloride profiles in tetrapods located in southern Brazil, obtained from samples extracted at 5 and 9.5 years from four microenvironments To this end, the equation of Fick's second lawconsiders the time-dependent Cs variation, a model was prepared for each exposure time profile measured from each microenvironment, which considered the Cs variation since it increased significantly from 5 to 9.5 years. The models obtained from the profiles measured at 5 years were used to estimate the profile at the exposure time of 9.5 years and then compared with the real profiles measured at 9.5 years. The same procedure was used for the profiles measured at 9.5 years, that is to say, a 5-year profile was estimated and compared with a profile measured at the same exposure time. This shows the efficiency of the model. According to the research results, profiles of about 5 years are still too premature to be used as an estimate of the residual useful life of a structure, even considering the Cs variation. However, profiles of approximately ten years can be used, and their Cs values may increase. Therefore, research of the Cs values must continue to obtain the corresponding older chloride profiles to assess the behavior of the curves and the efficiency of the suggested model.

Keywords: Durability, marine environment, chlorides, concrete, diffusion.

Introduction

Some studies (Rungrawee & Tetsuya, 2017; Meira et al., 2007; Ann et al., 2009; Song et al., 2008; Andrade et al., 2015) have demonstrated an increase in chloride deposition rates on the concrete surface (Cs) over time. Meira (2004) and Lu Feng (2017), who studied this rate over months, reported that this behavior is often conditioned by the types of marine atmospheres where the concrete structure is located. For the above, it is essential to study the effect of Cs on the chloride ion penetration rate due to the fact that the higher the Cs value, the greater the penetration rate into the concrete, which may lead to inevitable damage to the reinforcement concrete structure due to the corrosion of steel bar (Pazini, 2015). According to Mendoza et al. (2015) and Rincón et al. (2004), chloride deposition plays an important role, as an environmental indicator in the service life of concrete structures, in marine environments. Furthermore, some studies have shown that the Cs value tends to increase with exposure time (Silva, 2010). Accordingly, Guimarães et al. (2007) and Pereira (2003) report that for offshore structures of more than 20 years, the Cs tends to increase because of the exposure time of the concrete, but tends to stabilize when it reaches a specific maximum value. Vera et al. (2004) observed Cs variations proportional to the approximate square root of time. Likewise, Crank (1975) presented a model that uses the Cs variation and evaluate the model in which the Cs value varies, depending on the square root of time for the environment under study.

The present research exhibits the dynamics of chloride ion penetration into the concrete over time in tetrapods located in the eastern bar of the jetties to access the port complex in Rio Grande–RS-Brazil. To this end, the behavior of chloride profiles was monitored in December/2002 (5 years of exposure time) and in June/2007. These profiles were treated, and chloride penetration models were used by considering the concrete surface variation. Next, the results obtained from the model were compared with the profiles of tetrapods measured *in situ* at the exposure time under study, and the efficiency of the model was evaluated.

Description of the problem

Plenty of steel-reinforced concrete structures is constructed under specific chloride environments, such as marine/coastal environment, deicing salt environment, industrial wastewater environment, etc. Under such environments, chloride ions can easily penetrate into the concrete, since it is a porous material that has a porous binder phase and imperfect interfaces (Ma et al., 2014). Some studies have shown the increasing chloride content of concrete surface (Cs) that have been exposed to aggressive marine environments over time. In real concrete structure, chloride ions from the service environment can penetrate concrete and deposit in the surface layer, to form the boundary condition for further diffusion towards the interior. The deposit amount of chloride ions in the surface layer is usually a function of time, rather than a constant. According to Fick’s second law, the ingress of chloride ions is governed by two factors, i.e., the surface chloride concentration and the chloride diffusion coefficient of the matrix (Jun et al., 2014).

State of the Art

Chloride penetration models considering constant or variable Cs

Equations 1 and 2 presented by Crank (1975) are given as a solution of Fick's second law considering the diffusion coefficient (D), and considering the chloride content at the concrete surface (Cs) is constant.

$$C(x,t) = C_0 + (C_S - C_0).\operatorname{erfc}\left(\frac{x}{(\sqrt{4.D.t})}\right) \qquad (1)$$

where:
$C(x,t)$ = chloride ion concentration in relation to the cement mass at depth x from the surface of the concrete at time t (%);
C_0 = initial chloride ion concentration in the concrete (%);
C_S = surface concentration of chloride ions, accepted as constant (%);
x = depth of chloride ion penetration (mm);
D = diffusion coefficient of chloride ions (mm^2/year), accepted as constant;
t =time (years);
erfc = complementary Gaussian error function.

$$M_t = 2Cs\left(\frac{D_{Csconst}t}{\pi}\right)^{1/2} \qquad (2)$$

where:
M_t = total chloride mass which undergoes diffusion at time t (%.mm);
$D_{Csconst}$= diffusion coefficient of chlorides in concrete for constant Cs (mm^2/year);

Considering the Cs variation according to the square root of time (equation 3), equations 4 and 5 are presented, as given by Crank.

$$C_{x=0} = Cs = kt^{1/2} \qquad (3)$$

where:
Cs = chloride ion concentration on concrete surface – varies according to time (%);
k = parameter that considers Cs variation (%. year$^{-1/2}$);
t = concrete exposure time to chlorides (years).

$$\mathrm{Mt} = \frac{1}{2}\mathrm{kt}(\pi \mathrm{D}_{\mathrm{Csvar}})^{1/2} \tag{4}$$

where:
D_{Csvar} = diffusion coefficient considering Cs variation over time (mm^2/year).

$$\mathrm{C_x} = \mathrm{kt}^{1/2}\left\{\exp\left(-\frac{\mathrm{x}^2}{4\mathrm{D}_{\mathrm{Csvar}}\mathrm{t}}\right) - \frac{\mathrm{x}\pi^{1/2}}{2\sqrt{\mathrm{D}_{\mathrm{Csvar}}\mathrm{t}}}\mathrm{erfc}\frac{\mathrm{x}}{2\sqrt{\mathrm{D}_{\mathrm{Csvar}}\mathrm{t}}}\right\} = \mathrm{k}(\pi \mathrm{t})^{1/2}\mathrm{ierf}\frac{\mathrm{x}}{2\sqrt{\mathrm{D}_{\mathrm{Csvar}}\mathrm{t}}} \tag{5}$$

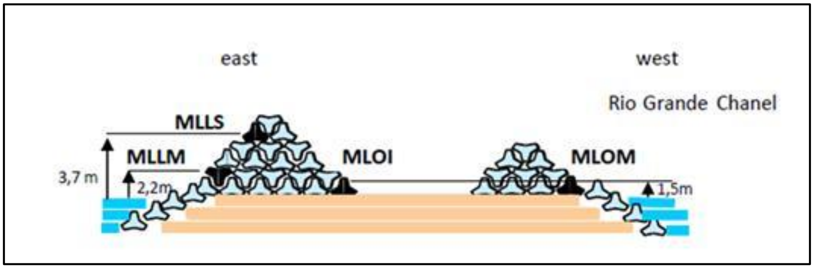
Equalizing the Mt values of equations 2 and 4, it is seen that for the same chloride profile, the diffusion coefficient is 62% higher when considering the constant Cs than that of the changing Cs, according to the square root of time. However, there is a significant difference between the two models.

Methodology

Four microenvironments of the eastern jetty were studied (Figure 1), located at the entrance of the Channel leading into the Rio Grande port complex, in RS-Brazil. Each of the microenvironment studied was represented by two tetrapods, totaling eight, using the following denominations:

- MLLS – microenvironment of the eastern jetty, facing east and at a higher elevation – 3.7 m above the seawater;
- MLLM – microenvironment of the eastern jetty, facing east and at an average elevation – 2.2 m above the seawater;
- MLOM – microenvironment of the eastern jetty, facing west and at an average elevation – 1.5 m above the seawater;
- MLOI – microenvironment of the eastern jetty, facing west and inside the jetty – 1.5 m above the seawater.

Figure 1 Location of microenvironments in the eastern jetty of the Rio Grande bar. Source: self elaboration.



The materials used to prepare the concrete were: coarse aggregate consisting of granite stone and fine aggregate consisting of quartz sand. Cement used was a High-early strength and sulfate resistant with fly ash containing 12%, with a compressive strength of 48 MPa at 28 days. The water used in the concrete had the following characteristics: chloride content of approximately 430 mg/l, sulfate content of 100 mg/l, pH of 7.3, solid residue of 250 mg/l, and O_2 consumption for the organic matter of 1.2 mg/l. According to the work reports, no additives were used. The average w/c ratio used for the concrete dosages was 0.5, and the results of compressive strength at 28 days (f_{ck}) was of approximately 32 MPa (Vera et al., 2014).

The samples used in the present research were obtained using a special drill, and the material was extracted in 5 mm layers from the outer surface to a depth of 50 mm. The sample consisted of the same-layer materials of two tetrapods, and the result represents the average chloride content for each microenvironment studied. From the contents of these samples, obtained at the exposure times of 5 years and 9.5 years, the chloride profiles were outlined.

After the chloride measurement profiles were outlined, using Equation 1, the regression of the chloride profiles was obtained using the minimum squared error method, with this equation relative to Fick's second law, considering constant Cs. It should be noted that this equation was only used to obtain a better definition of the measurement profiles and to estimate the Cs value at the exposure times studied.

The results showed a large Cs variation over time, which was fitted, approximately, according to the square root of time, and parameter k was estimated using Equation 3. Calculating the area under each regression curve yielded the Mt values and using equation 4 the D_{CSvar} was calculated for each exposure time studied. Equation 5 was used to obtain the estimated profiles at different exposure times.

According to results obtained by Guimarães et al. (2007) and Pereira (2003), a maximum Cs value of 3.5% was used for the tetrapods in relation to the cement mass. This equates to approximately 0.6% ofthe mass of concrete researched. Next, an estimate of the chloride profiles was prepared for exposure time at which it is assumed that the concrete has already reached the maximum value, and these curves were used to estimate the profiles at higher exposure times. The time to reach this maximum Cs was called the time change (t_{change}), that is, when the change occurs in the model considering the C_S variation (C_{Svar}) for constant C_S (C_{Sconst}) (Figure 2). To obtain each profile at each time change an equivalent time (t_{eq}) was calculated, which is the time required to achieve the profile at the time change considering Cs as constant. Therefore, the parity between equations 2 and 4 yielded Equation 6 (Figure 2).

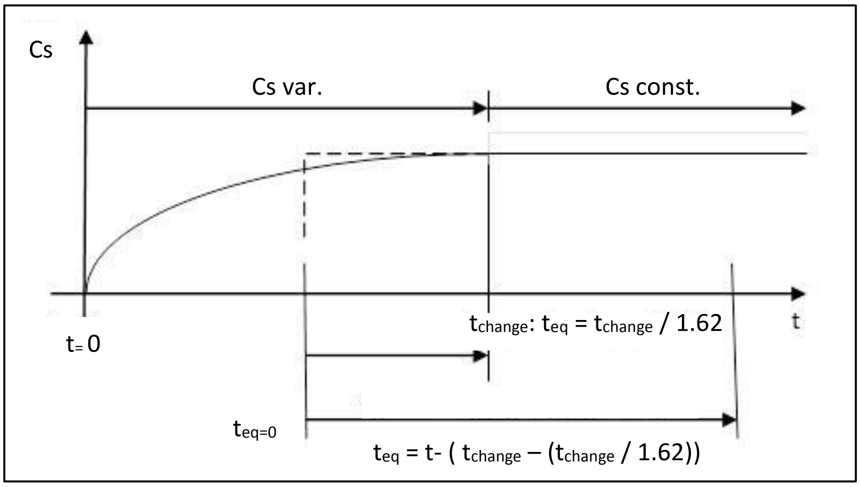
$$t_{change} = 1.62.\, t_{eq} \qquad (6)$$

where:

t_{change} = time for which C_S is maximum, from which C_S is considered constant and equal to 0.6% relative to the concrete mass;

t_{eq}= time for fitting the profile considering the C_S variation and profile considering the C_S constant.

Figure 2 Cs behavior over time. Source: Guimaraes (2007).



For higher exposure times t_{change} was calculated as time equivalent (t_{eq}’), according to Equation 7 and Figure 2.

$$teq' = t - (t_{change} - \left(\frac{t_{change}}{1.62}\right) \qquad (7)$$

where:

t_{eq}’ = equivalent time for t >t_{change}

All procedures described above were performed for the profiles measured at 5 years, and for the profiles measured at 9.5 years, considering a k for each exposure time studied. Furthermore, a k_{eq} was estimated through regression, which demonstrated the lowest squared error in the estimate of C_S variation between the exposure times studied. Therefore, with keq the same procedures used with k of 5 years and 9.5 years were repeated.

Using the model obtained for the 5-year profiles, the profiles at 9.5 years were estimated, which were compared with the chloride profiles measured at 9.5 years. The same procedure was performed with the model obtained for profiles at 9.5 years, in other words, from these curves the 5-year profiles were estimated, which were then compared with the profiles measured at this exposure time. This enabled to verify the efficiency of the model used.

In order to compare the performance of each model obtained a relative error was defined as:

$$E_{\text{Relative}} = \frac{E_{\text{Est.}}}{E_{\text{Min Reg.}}} \quad (8)$$

where:

$E_{\text{Est.}}$ = Square error of the estimate (between points measured and points estimated by the model);

$E_{\text{Mín.Reg.}}$ = Least square error of the regression (between points measured and points of its regression);

E_{relative} = Relative error.

Therefore, on the excellent performance of the model proposed this should present a small relative error.

Results

Figures 3/4/5 and 6 show the chloride profiles for the microenvironment MLLS, whose estimates showed the best accuracy in general, as will be seen later. Figures 7/8/9 and 10 illustrate profiles for microenvironment MLOM, estimates that have lower accuracy in general, as will be seen below.

Figure 3 Profiles of Cl^- for the model at 5 years ($k_{5\ years}$), for MLLS. Source: self elaboration.

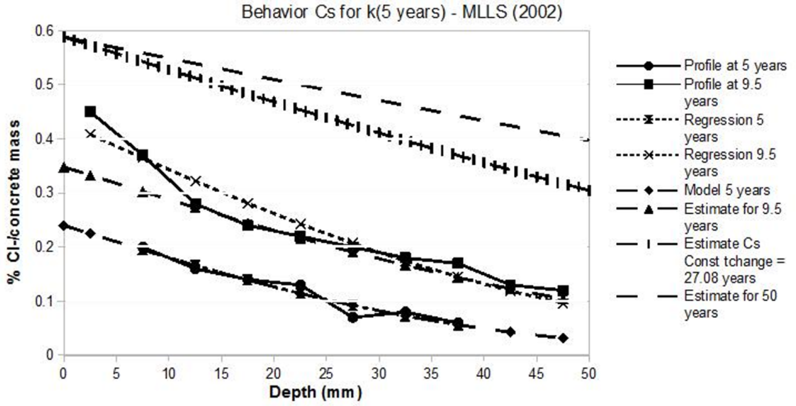


Figure 4 Profiles of Cl^- for the model at 9.5 years ($k_{9.5\ years}$), for MLLS. Source: self elaboration.

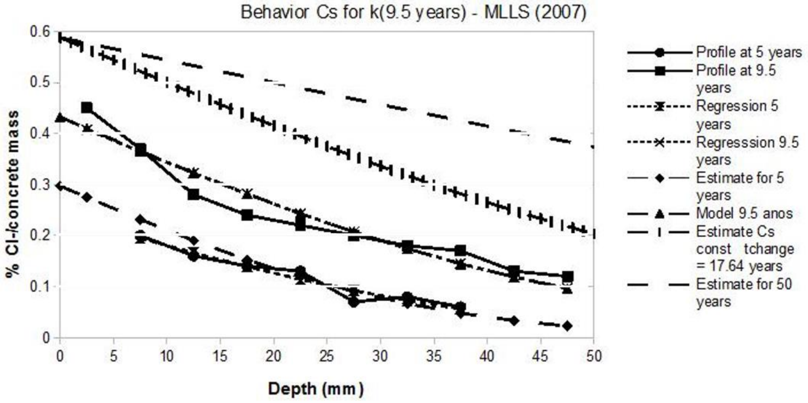


Figure 5 Profiles of Cl^- for the model at 5 years (k_{eq}), for MLLS. Source: self elaboration.

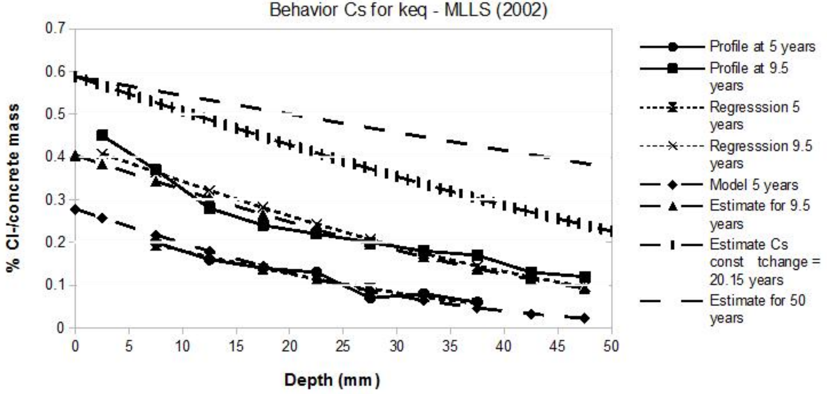


Figure 6 Profiles of Cl^- for the model at 9.5 years (k_{eq}), for MLLS. Source: self elaboration.

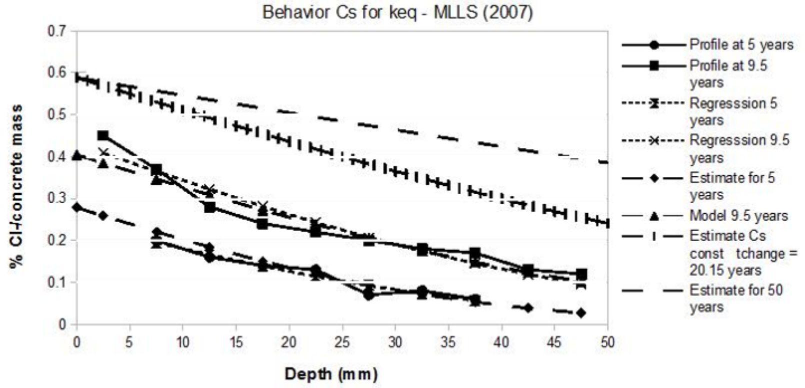


Figure 7 Profiles of Cl^- for the model of 5 years (k_{5anos}), for MLOM. Source: self elaboration

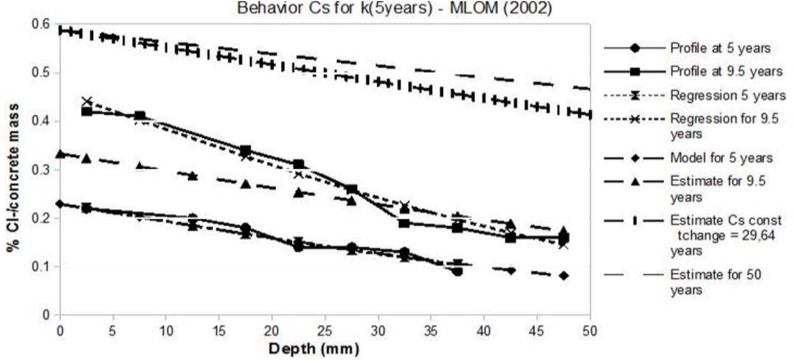


Figure 8 Profiles of Cl^- for the model of 9.5 years (k_{5anos}), for MLOM. Source: self elaboration.

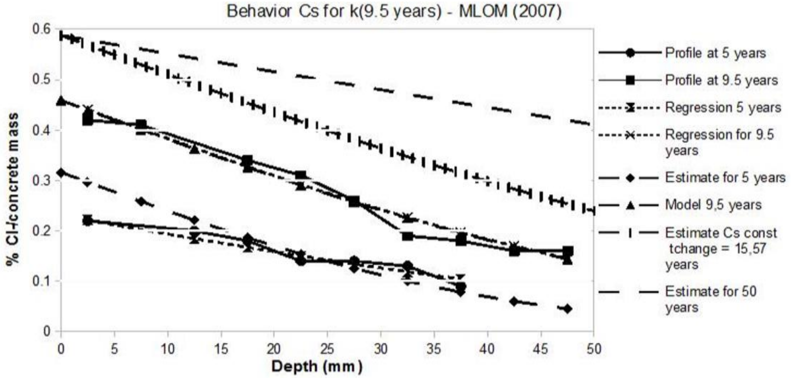


Figure 9 Profiles of Cl^- for the model of 5 years (k_{eq}), for MLOM. Source: self elaboration.

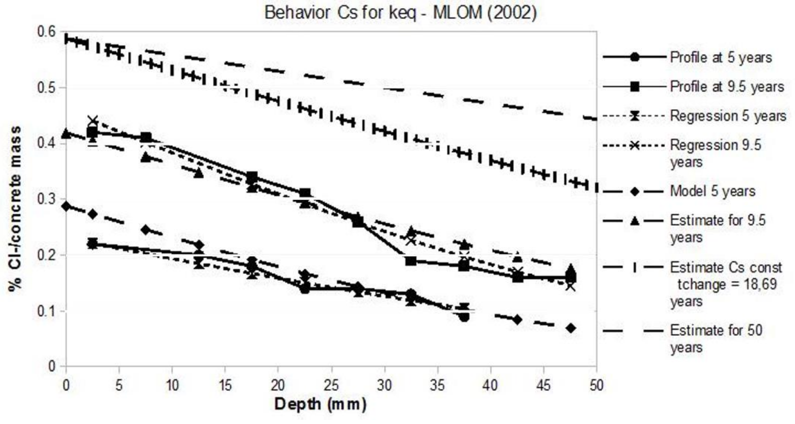


Figure 10 Profiles of Cl⁻ for the model of 9.5 years (k_{eq}), for MLOM. Source: self elaboration.

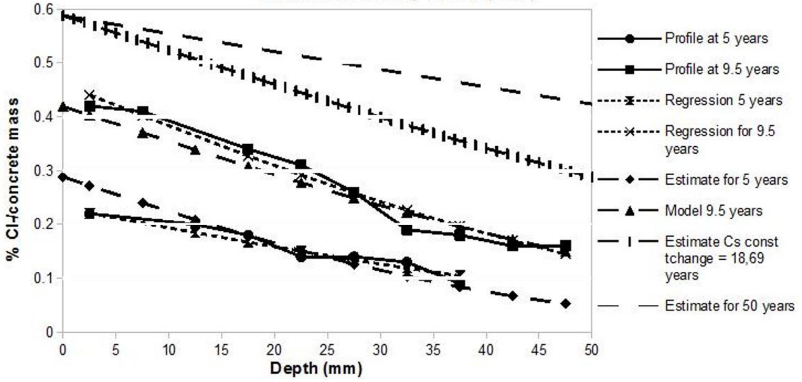


Table 1 shows the estimated parameters for the 4 micro-environments studied.

Comparing the $E_{relative}$ values with the curves obtained through the model, the following classification was used to better analyze the results: between 1 and 2 – very good (VG); between 2 and 4 – good (G); between 4 and 10 – regular (R); and greater than 10 – poor (P).

Table 1. Estimated parameters for the 4 microenvironments of the tetrapods under study. Source: self elaboration.

			5years			9.5 years			*5years			*9.5years		
Micro environments	Cs_{5years} %	$Cs_{9.5years}$ %	D (mm²)	t_{change} (year)	$E_{relative}$ year	D (mm²)	t_{change} (year)	$E_{relative}$ year	D (mm²)	t_{change} (year)	$E_{relative}$ year	D (mm²)	t_{change} (year)	$E_{relative}$ year
MLLS	0.24	0.43	179	27	2.7	129	18	3.4	133	20	1.2	147	20	2.2
MLLM	0.34	0.59	593	14	22.9	452	9	3.9	465	11	3.6	503	11	2.4
MLOM	0.23	0.46	470	30	10.7	189	16	9.1	296	19	2.8	227	19	4.4
MLOI	0.43	0.44	185	8	11.4	399	17	1.4	291	13	2.5	311	13	1.1

Key:* Classification of $E_{relative}$ - 1 to 2 – MB 2 to 4 – B 4 to 10 – Reg >10 – P *Considering k_{eq}

Discussion

The analysis of the measurement profiles at 9.5 years showed better defined curves, as the five-year profiles also showed peaks or oscillations that disappeared at the exposure time of 9.5 years. The better-defined profile at 5 years was the microenvironment MLLS, which showed no peak or oscillation at that exposure time and demonstrated a very close behavior in the chloride profiles at 5 years and at 9.5 years, and the microenvironment MLLM exhibited a small peak at five years. But the microenvironment MLOM exhibited oscillation in the 5 years profile. The MLOI point showed a e large chloride content at a considerable depth, which was 100 mm. According to the limits of chloride concentration exhibited by ACI 318 and considering the characteristics of concrete used in the present work, chloride content of 0.08 % relative to the concrete mass, generates the depassivation, in the case of structure will use reinforcing steel (profile at 9.5 years).

In the four microenvironments studied, the chloride content at the concrete surface (Cs) increased from 5 to 9.5 years, and the diffusion coefficient (D) decreased from 5 to 9.5 years. This was expected because the Cs values tend to increase with time until it reaches a certain maximum value. According to Table 1, this behavior is evident, except for MLLM, which could have already reached the fixed maximum chloride content of 0.6 % relative to the concrete mass, a typical value for structures with more than 20 years of exposition, as proposed by Guimarães et al. (2007) and Pereira (2003).

Overall, the chloride content at the concrete surface – Cs – increased with time (Table 1). However, the 9.5-year profiles were better defined because the 5 year profiles still showed peaks or oscillations which disappeared at 9.5 years. This could be the reason why some microclimates exhibit considerable differences between the profiles estimated at 9.5 years (when using the model based on the profiles measured at 5 years) and the profiles measured at 9.5 years. At the exposure time, when the Cs value is higher, the chloride attack depth in the concrete is also higher, which is consistent with the useful life prediction models (Silva, 2014).

According to the results obtained in this study, profiles of approximately five years are still too premature to be used as an estimate of the residual service life of a concrete structure, even considering Cs variation. However, profiles of approximately 10 years can be used, but it should be anticipated that over time their Cs values may increase. Overall, there was a better performance of the estimated profiles when using a keq than when using a k of that exposure time. This was confirmed by the results obtained in Figures 3 and 6, where estimates at 9.5 years were somewhat distant from the measurement profiles at that exposure time. The same was confirmed through the $E_{relative}$ values (Table 1), where the worst estimates were for the 5 year profiles, except for MLLS which showed good estimate at 9.5 years. It is observed that the 5 year estimate profiles obtained from profiles measured at 9.5 years of exposure time for both k and keq showed, in general, higher similarity – due to the curves' behavior (Figures 4, 6, 8 and 10); or from the results shown in Table 1, where the 4 microenvironments studied exhibited $E_{relative}$ from good (G) to very good (VG), except for MLOM which exhibited regular (R) $E_{relative}$. The analysis of the chloride profiles clearly showed that profiles at 9.5 years exhibited better-defined curves than the profiles at 5 years.

Furthermore, not only the importance of the model was verified, but also the aggressiveness of the environment studied. It is also important to mention that according to the results obtained in the present research, Guimarães (2000) and Rodrigues (2009) the high-early strength and sulfate resistant cement, with 12% of fly ash, did not present convenient result to be used in constructions located in marine environments. As an example, we mention the microenvironment MLOM, which exhibited high chloride content at a considerable depth in only 5 years of concrete exposure, which may have caused premature steel depassivation, if reinforced concrete was used instead of mass concrete.

This study should be continued, obtaining chloride profiles at older exposure time, that is, tetrapods with 15 or 20 years of exposure, in order to assess the behavior of the curves and the efficiency of the model used in this work. Additionally, verifying whether the Cs values have also increased with time, or have already reached the maximum value suggested. It should be emphasized that Crank's equation is particularly applicable to assess concrete structures at short exposure time when the Cs variation has a significant influence on the concrete structure.

Conclusions

Considering the chloride content variation at the concrete surface, Crank's model showed good results to estimate the chloride profiles. Different from that demonstrated by Silva and Guimarães (2000), Equation 5 showed the expected behavior, with a reduction in chloride content from the surface to the interior of the concrete.

The analysis of the chloride profiles showed that profiles at 9.5 years had better-defined curves than profiles at 5 years. Therefore, profiles of approximately 5 years of exposure time are still too premature to be used to estimate the residual service life of a structure, even considering the Cs variation. Therefore, profiles of approximately 10 years can be used; however, keeping in mind that their Cs values may increase over time.

Additionally, not only was the importance of the model verified, but also the aggressiveness of the environment studied. An example worth mentioning is the microenvironment MLOM, which exhibited profiles with high chloride content at a considerable depth, and with only five years of exposure, which could have caused premature steel depassivation, if reinforced concrete was used. Overall, better performance of the estimated profiles was obtained when a keq was used (coefficient of variation of the Cs) than when the "k" of that exposure time was used.

The importance of continuing this research is underscored, that is, to obtain chloride profiles at higher exposure time, not only in the structure investigated in this study, but also in other structures in order to assess the behavior of the curves and the efficiency of the model used, and also to investigate the Cs values, if they increase with exposure time or may have already reached the maximum suggested.

Acknowledgement

We are grateful to Fundação de Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - CAPES for the financial to perform this work. The study was conducted in the city of Rio Grande, Rio Grande do Soul, Brazil.

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