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Engineering expression of the ClinConc model for prediction of free and total chloride ingress in submerged marine concrete

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ABSTRACT

This paper intends to express the physical model ClinConc in a more engineer-friendly way. Through the numerical evaluation it has been found that there exist good correlations between the time-dependent factors for chloride binding and for diffusion coefficient, and between the laboratory measured diffusion coefficient and the apparent one, the latter is a result of the combined effects of material properties and environmental actions. Therefore, the ClinConc model can be expressed by the similar error function as used in many empiric models, but with the proper physical procedures, that is, modeling free chloride transport taking free chloride as diffusion potential and then calculating the total chloride content taking into account the non-linear chloride binding. The modeled chloride profiles are in good agreement with the measured ones, even though further study is needed to clarify the expansion coefficient for the concrete containing silica fume and other pozzolanic additions.

1. Background

The models for prediction of chloride ingress in concrete can be roughly divided into two groups: empiric and physical models. Empiric models “empirically” take the total (or acid soluble) chloride as driving potential, which is, strictly speaking, not correct, because it is free chloride that has the electrochemical potential for diffusion. Therefore, physical models should be based on the physical and chemical processes involved in the chloride transport in concrete, for instance, free chloride as driving potential, chloride binding, convection, multi-ionic characteristics, etc. The conventional ERFC (ERror Function Complement) based models as recently reviewed by Nilsson and Carcasses [1] are typically empiric ones due to their assumption of total chloride as driving potential. These empiric models are widely used by engineers and researchers in practical applications, probably because of their relatively simple mathematical expressions when compared with physical models that often need sophistical calculations and iterations due to the lack of analytical mathematical solutions. There are a number of physical models [2–6]. One of them is the ClinConc model, which was developed in the middle of 1990s [2] and consists of two main procedures:

1) Simulation of free chloride penetration through the pore solution in concrete using a genuine flux equation based on the principle of Fick's law with the free chloride concentration as the driving potential, and
2) Calculation of the distribution of the total chloride content in concrete using the mass balance equation combined with non-linear chloride binding.

Obviously, the ClinConc model uses free chloride as the driving force and takes non-linear chloride binding [7] into account. Thus it describes chloride transport in concrete in more scientific way than the empiric models. On the other hand, since the ClinConc model needs numerical iterations for the binding effect, its practical engineering applications have been restrained due to the need of computation for numerical iterations. Therefore, it is necessary to simplify the model using more engineer-friendly expression.

2. Time-dependent chloride diffusion coefficient

Very recently, Tang and Gulikers [8] verified the mathematics of time-dependent chloride diffusion coefficient and gave the more correct expression as follows:

$$\frac{c}{c_s} = 1 - \operatorname{erf}\left(\frac{x}{2\sqrt{D_a \cdot t}}\right) \quad (1)$$

where c and c_s denote the concentration of dissolved (free) chloride in the pore solution within the concrete cover and at the exposed concrete surface, respectively, erf is the error function, x is the distance, t is the duration of chloride exposure, and D_a is referred to as

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the apparent chloride diffusion coefficient that is not a constant but time-dependent:

$$D_a = \frac{D_0}{1-n}\cdot\left(\frac{t'_0}{t}\right)^n\cdot\left[\left(1+\frac{t'_{ex}}{t}\right)^{1-n}-\left(\frac{t'_{ex}}{t}\right)^{1-n}\right] = D_0\cdot f(n,t) \quad (2)$$

where D_0 is the diffusion coefficient of Fick's 2nd law at age t'_0, n is the age factor, and t'_{ex} is the age of concrete at the start of exposure. Notice that, in this paper t' denotes the age of concrete and t denotes the duration of exposure.

From the measurement on young concrete at the early age without previous chloride exposure, Tang and Nilsson [9] found that the measured chloride diffusion coefficient decreased with age. After a certain age, like the development of hydration process, the chloride diffusion coefficient becomes more or less constant according to Tang's laboratory test [10]. Therefore, in the original ClinConc model the time-dependent factor of chloride diffusion coefficient, $f_D(t')$, is described by the following equation [2]:

$$f_D(t') = \begin{cases} \left(\frac{t'_{D_s}}{t'}\right)^{\beta_t} & t' < t'_{D_s} \\ 1 & t' \geq t'_{D_s} \end{cases} \quad (3)$$

where t'_{Ds} is the age when the diffusion coefficient becomes stable and β_t is a constant (age factor for young concrete). The age t'_{Ds} is normally about 0.5 year for ordinary cement concrete and perhaps a longer time for some concrete containing pozzolanic additions. This age is relatively short when compared with the service life of concrete structures. However, it has been found from the field chloride profiles that the apparent chloride diffusion coefficient curve-fitted to the ERFC solution to Fick's 2nd law, decreases even after very long exposure time [11–14]. One of the explanations to the decrease in curve-fitted D_a is the continuous densification of concrete [1], but so far no convincing evidences show this densification effect, because if it is true, the conductivity of concrete should be proportionally decreased, keeping in mind that the conductivity of chloride ions is less than that of hydroxide ions. In contrast, some field investigations show that the high strength concrete become more porous after field exposure [15,16]. Obviously, there must be some other reasons to the decrease in the curve-fitted D_a. It will be shown later that the decreased apparent diffusion coefficient is, to a great extent for the chloride ingress under submerged conditions, attributed to a time-dependent chloride binding.

3. Time-dependent chloride binding

From observations, it has been found that the total chloride content at the surface of concrete, C_s (notice the upper-case for chloride content in concrete), also increases with exposure time [17–20], even for concrete exposed under submerged conditions [18–20], where the free surface-chloride concentration c_s (notice the lower-case for chloride concentration in solution) remains relatively constant over time. Tang and Nilsson [19] attributed the increased surface-chloride content C_s under submerged conditions to the increased chloride binding capacity and proposed the following equation to describe the time-dependent factor for chloride binding, f_t:

$$f_t = a_t \ln(t_{Cl}+0.5)+1 \quad (4)$$

where t_{Cl} is the chloride contamination duration in years and a_t is a constant. According to the data from over 10 years field exposure in Swedish marine environment [20], the typical value of a_t is 0.36, but may vary from 0.1 to 0.6 depending on binder type, pozzolanic addition and water–binder ratio.

For the chloride ingress in splash and atmospheric zones, the increased concentration of free chlorides due to drying process may be another important reason, which is out of the scope of this paper.

4. Relation of the apparent diffusion coefficient to the laboratory measured one

The diffusion coefficient D_0 in Eq. (2) can be considered as an "initial" apparent one, which is a result of the combined effect of transport property of concrete material and environmental actions. A diffusion coefficient measured in the laboratory reflects more or less the transport property under standard test conditions. In the ClinConc model, the coefficient measured at an age of 6 months using the Nordic standard rapid migration test NT BUILD 492, denoted as D_{6m}, is used as an input constant value. Basically, the diffusion coefficient D_0 and the migration coefficient D_{6m} should have the following relationship [2]:

$$D_0 = \frac{1+0.59K_{b6m}}{1+\frac{\partial c_b}{\partial c}}\cdot D_{6m}\cdot k_{TD} \quad (5)$$

where c_b is the bound chloride, $\frac{\partial c_b}{\partial c}$ is the chloride binding capacity (or more correctly wording, chloride binding rate), k_{TD} is the temperature factor for diffusion coefficient, and

$$K_{b6m} = \frac{W_{gel6m}}{1000\varepsilon_{6m}} \quad (6)$$

where W_{gel} is the gel content in kg/m$^3_{concrete}$ and ε is the water accessible porosity. The subscript "6m" denotes the value at the age 6 months. According [2],

$$\frac{\partial c_b}{\partial c} = k_{OH}\cdot k_{Tb}\cdot\frac{W_{gel}}{1000\varepsilon}\cdot f_b\cdot\beta_b\cdot c^{\beta_b-1} \quad (7)$$

where k_{OH} is the factor describing the effect of alkalinity, k_{Tb} is the temperature factor for chloride binding, f_b and β_b are chloride binding constants. Calculations of the above parameters as well as the related hydration degree α_h have been given in [2]. Since the free chloride concentration c is depth-dependent, the binding rate in Eq. (7) cannot be directly obtained. On the other hand, the time-dependent chloride binding will also influence the relationship shown in Eq. (5). Therefore, it is reasonable to consider the chloride binding at the surface of concrete under the exposure conditions of c_s and T, and add a new factor Y to bridge the gap between D_0 and the migration coefficient D_{6m}.

$$Y = \frac{1+k_{OH6m}\cdot K_{b6m}\cdot k_{Tb}\cdot f_b\cdot\beta_b\cdot\left(\frac{c_s}{35.45}\right)^{\beta_b-1}}{(1+0.59K_{b6m})\cdot k_{TD}}\cdot\frac{D_0}{D_{6m}} \quad (8)$$

with

$$k_{OH6m} = e^{0.59\left(1-\frac{0.043}{[OH]_{6m}}\right)} \quad (9)$$

$$k_{TD} = e^{\frac{E_D}{R}\left(\frac{1}{T_0}-\frac{1}{T}\right)} \quad (10)$$

$$k_{Tb} = e^{\frac{E_b}{R}\left(\frac{1}{T}-\frac{1}{T_0}\right)} \quad (11)$$

where $[OH]_{6m}$ is the hydroxide concentration in mol/dm^3 at the age of 6 months, E_D and E_b are the activation energy of diffusion coefficient and chloride binding, respectively, and T_0 is the temperature in the

Table 1
Mixture proportions of concrete

Mix no.	Binder type	Binder content kg/m^3	w/b	Air content %	α_{h6m}	W_{gel6m} kg/m^3	ε_{6m}	[OH]$_{6m}$ mol/l
1	SRPC	490	0.30	3.6	0.5	310	0.11	0.73
2	SRPC	450	0.35	6	0.63	350	0.11	0.68
3	SRPC	420	0.40	6	0.75	390	0.11	0.53
4	SRPC	370	0.50	6	0.85	390	0.13	0.53
5	SRPC	240	0.75	6	0.96	290	0.14	0.41
6	95% SRPC+5% silica fume (SF)	420	0.40	6	0.75	390	0.11	0.25
7	82% SRPC+4% SF+14% flyash (FA)	420	0.40	6	0.76	360	0.11	0.79
8	OPC	420	0.40	6	0.75	390	0.11	1.31

laboratory conditions. At the exposure surface, the hydroxide concentration is assumed to be 0.043 mol/dm^3, as in the saturated $Ca(OH)_2$ solution.

Thus it is possible to evaluate the factor Y by means of the ClinConc model with numerical calculations and the curve-fitting technique to obtain apparent diffusion coefficient D_0, as to be presented in the next chapter.

5. Numerical evaluation

The evaluation will be limited to the submerged condition with concrete age at start of exposure t'_{ex}=0.5 month (0.04 year). Thus the parameters influencing chloride transport in a homogeneous concrete (excluding the skin-effect) will be as follows:

- Chloride diffusion coefficient measured by the standard migration method (NT BUILD 492) at the age 6 months, D_{6m} (assuming t'_{Ds}=0.5 year);
- Chloride binding constants, f_b and β_b;
- Time-dependent factor for chloride binding, a_t;
- Environmental parameters, c_s and T.

The age factor for young concrete, β_t, as expressed in Eq. (3), was not included in the evaluation, because according to the previous evaluation [21], if t'_{Ds} in Eq. (3) is about 0.5 year, the parameter β_t has only limited effect on chloride, while excluding this factor can greatly simplify the mathematical expression without resulting in large difference in long-term prediction of service life.

Some typical mixture proportions of concrete as listed in Table 1 were used in the evaluation. The magnitude ranges of various parameters including the constants are listed in Table 2. As usual, the activation energies E_D and E_b are assumed as 42,000 and 40,000 J/mol, respectively. Although the values have not been validated yet, this assumption will not influence the evaluation.

The MS Excel based program version 4d, dated 2003 Oct., was used for calculation of chloride penetration profiles at the exposure times from 1 to 100 years. The predicted free chloride profiles were used for

Table 2
Magnitude ranges of input parameters

Parameter	Magnitude
Diffusion coefficient tested in the laboratory (e.g. according to NT BUILD 492 at the age t'_0 months and the temperature T_0=293 K), D_{6m}	1, 3, 6, 12×10^{-12} m^2/s
Chloride binding coefficient f_b	2.6, 3.6, 4.6, 6
Chloride binding exponent β_b	0.28, 0.33, 0.38, 0.43
Time-dependent factor of chloride binding, a_t	0–0.6 (0.36 was suggested in [19])
Chloride concentration in the seawater c_s	5, 14, 25 g/l
Water temperature, T	278, 284, 293 K (5, 11, 20 °C)

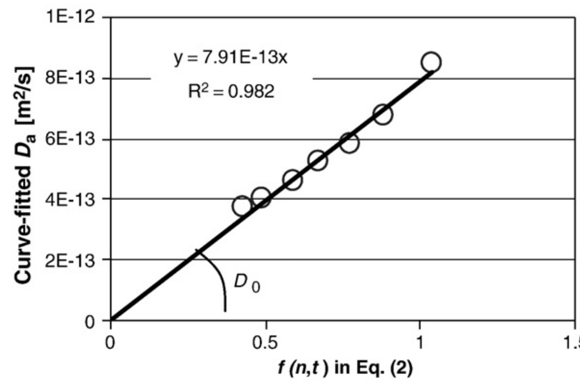

Fig. 1. Example of regression in order to obtain n and D_0 in Eq. (2).

curve-fitting to Eq. (1) in order to obtain D_a. Thus through the relationship between D_a and function $f(n, t)$ in Eq. (2) the best correlation can be achieved by adjusting the n value. The slope of the regression line with the best correlation is the value D_0, as shown in Fig. 1.

From the results the relationship between the n values and the time-dependent factor of chloride binding, a_t, can be obtained, as shown in Fig. 2, where the mean values of n and their standard deviation as error bars from various input parameters are used. Apparently, n is strongly related to a_t, irrespective of the other input parameters, such as D_{6m}, f_b, β_b, c_s and T as listed in Table 2, and the mixture proportions as listed in Table 1. It should be noticed that, in each modeling, the input diffusion coefficient, D_{6m}, was kept constant. The age factor n is, therefore, mainly attributed to the increase in chloride binding capacity through the parameter a_t, as can be expressed by the following regression equation:

$$n = -0.45a_t^2 + 0.66a_t + 0.02 \tag{12}$$

with a relatively constant standard deviation of 0.009.

The results also reveal that Eq. (8) has a very good correlation with the time-dependent factor of chloride binding, a_t, as shown in Fig. 3. Thus the initial apparent diffusion coefficient D_0 can be expressed by

$$D_0 = \frac{(0.8a_t^2 - 2a_t + 2.5)\cdot(1 + 0.59K_{b6m})\cdot k_{TD}}{1 + k_{OH6m}\cdot K_{b6m}\cdot k_{Tb}\cdot f_b\cdot \beta_b\cdot\left(\frac{c_s}{35.45}\right)^{\beta_b - 1}}\cdot D_{6m} \tag{13}$$

with a relatively constant COV (coefficient of variation) of 5.6%.

It can be seen from Figs. 2 and 3 or Eqs. (12) and (13) that the time-dependent factor for chloride binding, a_t, plays an important role in retarding chloride transport in concrete, in the form of a decreased apparent diffusion coefficient through the age factor n. All the three parameters of chloride binding a_t, f_b and β_b tend to decrease the ratio D_0/D_{6m}, but this decrease is not time-dependent. The apparent

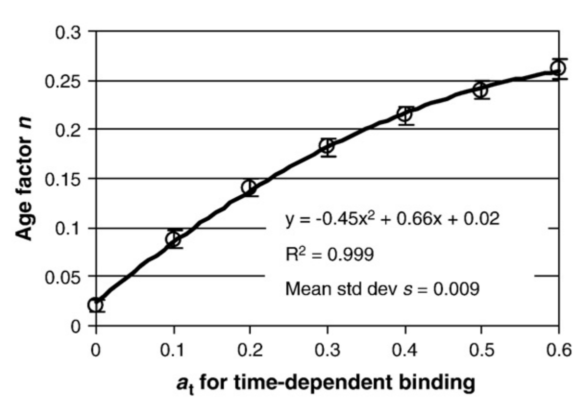

Fig. 2. Correlation between time-dependent factors n and a_t.

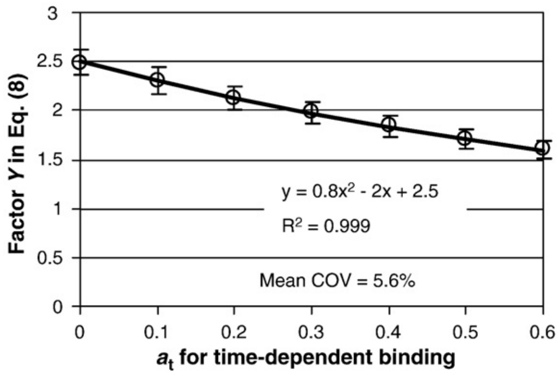

Fig. 3. Correlation between factor Y in Eq. (8) and time-dependent factor a_t.

diffusion coefficient D_0 is also dependent on exposure environment through the parameters c_s and T.

6. Engineering expression of the ClinConc model

6.1. Modeling of free chloride ingress

It can be seen from the above evaluation that it is the increased chloride binding which through the parameter a_t manifested a decreased apparent diffusion coefficient D_a. Since there are very clear correlations between n and a_t, and between D_0 and a_t, it is convenient to express the model in a more engineer-friendly way. Similar to the original principles of the ClinConc model, the first step is to model the free chloride ingress using the following equation:

$$\frac{c}{c_s} = 1 - \mathrm{erf}\left(\frac{x}{2\sqrt{\frac{k_D \cdot D_0}{1-n} \cdot \left[\left(1+\frac{t'_{ex}}{t}\right)^{1-n} - \left(\frac{t'_{ex}}{t}\right)^{1-n}\right] \cdot \left(\frac{t'_{6m}}{t}\right)^{n} \cdot t}}\right) \quad (14)$$

where n is as in Eq. (12), D_0 as in Eq. (13), and k_D is an extension coefficient.

From the ten years traceable investigation [20] it has been found that an extension coefficient k_D sometimes has to be added in the numerical simulation using the ClinConc model in order to obtain better agreement with the chloride profiles from the field exposure, especially in concrete with very low water–binder ratio or blended with puzzolanic additions. Possible reasons include:

- The laboratory test method may underestimate the diffusivity in concrete with very low water–binder ratio or blended with puzzolanic additions, due to the difficulty in fully saturating the test specimens;
- The actual diffusivity in the concrete exposed to the field may be higher than that cured under the laboratory condition, due to relatively worse curing conditions, e.g. impact of temperature and humidity cycles.

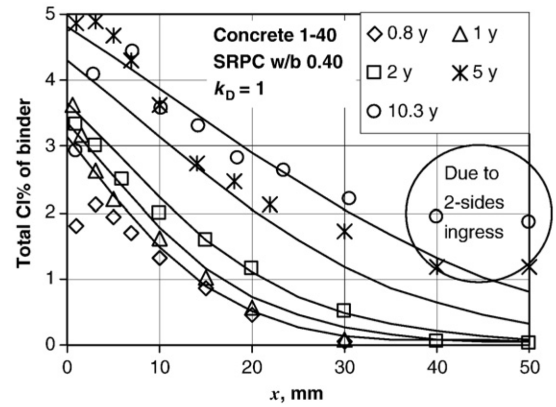

Fig. 5. Measured and modeled profiles for SRPC concrete w/b 0.40.

It has been shown from limited experimental data that, when using the same test method, the concrete taken from the field site indeed shows a diffusion coefficient higher than that cured under the laboratory condition [20,16]. Therefore, the extension coefficient k_D is for correcting possible difference in D_{6m} between the laboratory cured concrete and the field exposed one [20]. Further study is needed to find the real reasons, but this might be an indication that the diffusion coefficient does not decrease but increase with time.

If the diffusion coefficient is measured at the age $t'_{meas}<6$ months, the value of D_{6m} can be obtained by

$$D_{6m} = D_{meas} \cdot \left(\frac{t_{meas}}{t_{6m}}\right)^{\beta_t} \quad (15)$$

The β_t value can be estimated from a series of measurements at variable ages. As mentioned previously, the effect of β_t on long-term chloride ingress does not taken into account due to the short time of t_{D0} in Eq. (3) when compared with the service life of concrete structure. However, if the time t_{Ds} has been proven very long and its effect on long-term chloride ingress cannot be ignored, the value n in Eq. (12) then has to be modified.

If the threshold chloride concentration is expressed as free chloride concentration in the pore solution, or the ratio $[Cl^-]/[OH^-]$, the above equation is ready for use in service life calculation for corrosion initiation.

6.2. Calculation of total chloride content

If the threshold chloride concentration is expressed as total chloride content in concrete, further step is needed to convert the free chloride concentration to total chloride content. Basically, total chloride concentration c_{tot} is the sum of bound chloride, c_b, and free

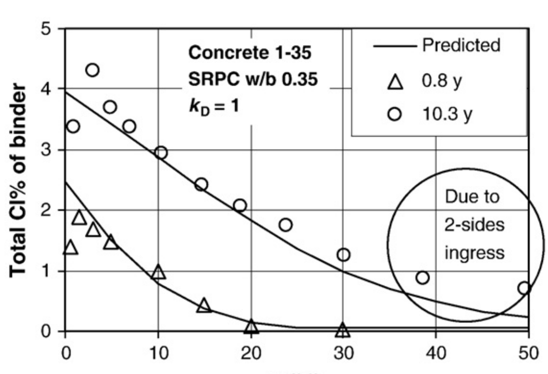

Fig. 4. Measured and modeled profiles for SRPC concrete w/b 0.35.

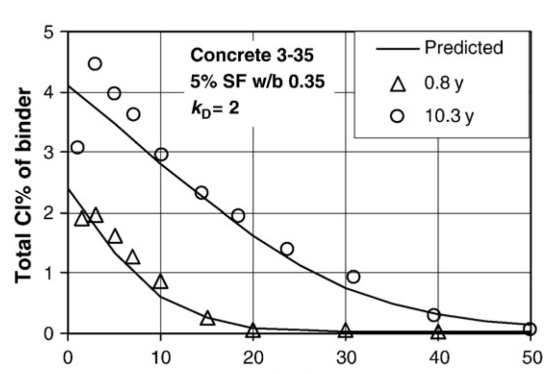

Fig. 6. Measured and modeled profiles for concrete containing 5% silica fume w/b 0.35.

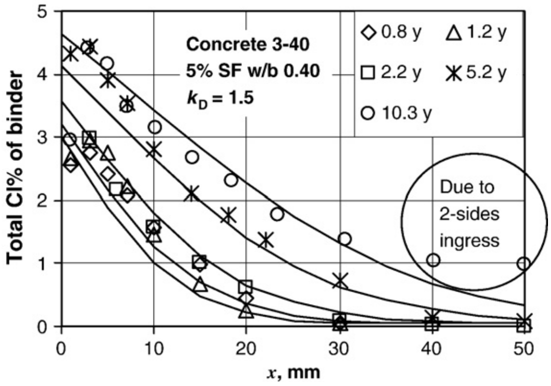

Fig. 7. Measured and modeled profiles for concrete containing 5% silica fume w/b 0.40.

chloride *c*. If the relationship between total and free chlorides is experimentally known, it should be no problem for the users of the ClinCon model to convert the predicted free chloride profiles as expressed in Eq. (14) to the total chloride profiles. Otherwise the following equation could be used [2]:

$$C = \frac{\varepsilon \cdot (c_b + c)}{B_c} \times 100 \text{ mass\% of binder} \tag{16}$$

where ε is the water accessible porosity at the age after the exposure, B_c is the cementitious binder content, in $kg/m^3_{concrete}$, and

$$c_b = f_t \cdot k_{OH6m} \cdot K_{b6m} \cdot k_{Tb} \cdot f_b \cdot c^{\beta_b} \text{ g/l} \tag{17}$$

According to Eq. (4), the time-dependent factor f_t is dependent on the chloride contamination time t_{Cl}. At the surface of concrete, $t_{Cl}=t$, while in the deep concrete where chlorides do not reach, $t_{Cl}=0$. Therefore, it is reasonable to express t_{Cl} as proportional to the free chloride concentration, that is, $t_{Cl} / t = c / c_s$. Thus f_t in Eq. (17) can be calculated by the following equation:

$$f_t = a_t \ln\left(\frac{c}{c_s} \cdot t + 0.5\right) + 1 \tag{18}$$

Now it can be seen that the ClinConc model has been expressed by the equations without need for iteration procedure. Thus it is no longer complicated when compared with other empiric models. The significant advantage of the ClinConc model is that it models the free chloride transport with clear physical and chemical meanings.

7. Some examples of modeling

Some examples of chloride profiles modeled basically using Eqs. (14) and (16) as the engineering expression of the ClinConc model, are shown in Figs. 4–8. The mixture proportions of concrete and input parameters used in the modeling are listed in Table 3. The concrete specimens were exposed in the seawater at Swedish west coast, where c_s = 14 g/l and T = 11 °C, the same as used in the previous publications [2,19,20,22].

It can be seen from the above examples that the modeled profiles are in good agreement with the measured ones, but with the expansion coefficient k_D = 1 for normal cement concrete, 1.5–2 for the concrete containing silica fume and 4 for the concrete containing blended fly ash and silica fume. This indicates that either the laboratory test method underestimated the value of D_{6m} in such types of concrete or the actual diffusion coefficient in such types of concrete increased during the field exposure — an agreement with the findings in [15,16,20,22].

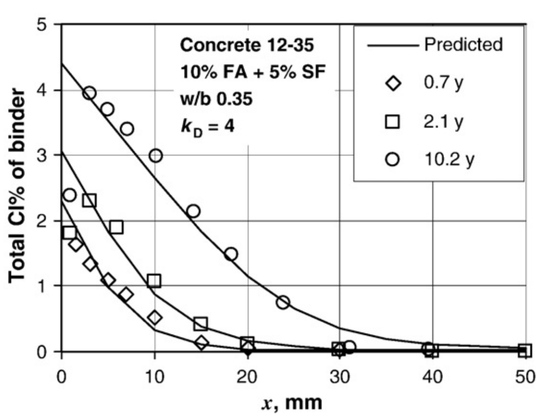

Fig. 8. Measured and modeled profiles for concrete containing 10% fly ash and 5% silica fume w/b 0.35.

Table 3
Mixture proportions of concrete and input parameters for modeling

Parameter	1–35	1–40	3–35	3–40	12–35
Binder	SRPC	SRPC	95% SRPC+ 5% SF	95% SRPC+ 5% SF	85% SRPC+ 5% SF+10% FA
B_c, kg/m^3	450	420	450	420	450
w/b	0.35	0.40	0.35	0.40	0.35
Air, %	6.0	6.2	5.8	6.1	6.4
D_{6m}, $\times 10^{-12}$ m^2/s	6.9	12.2	2.9	4.4	1.0
f_b	3.6	3.6	3.6	3.6	4.3
β_b	0.38	0.38	0.38	0.38	0.38
a_t	0.5	0.36	0.5	0.36	0.5
k_D	1	1	2	1.5	4

8. Concluding remarks

From the numerical calculation using the ClinConc model it has been found that the time-dependent chloride binding plays an important role in retarding chloride transport in concrete, in the form of a decreased apparent diffusion coefficient through the age factor *n*.

A good correlation between the time-dependent age factor *n* for diffusion and time-dependent factor a_t for chloride binding has been found. When the chloride binding parameters and the environmental parameters are taken into account, there also exists a good correlation between the initial apparent diffusion coefficient and the laboratory measured one. Thanks to these good correlations, the ClinConc model can be expressed in a more engineer-friendly way, that is, basically by Eq. (14) for free chloride distribution and Eq. (16) for converting free chloride to total chloride distribution. The modeled chloride profiles are in good agreement with the measured ones, even though further study is needed to clarify the expansion coefficient for the concrete containing silica fume and other pozzolanic additions.

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