CAPILLARY SUCTION AND DIFFUSION MODEL FOR CHLORIDE INGRESS INTO CONCRETE

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Abstract

A numerical approach, named TransChlor, is proposed to simulate transport phenomena of various substances in concrete. This approach is a theoretical model based on finite elements and finite differences methods. The model consists of coupled nonlinear partial differential equations based on Fick's diffusion law and on kinematics equations. Simulation results from a parametrical study highlight the influence of microclimatic conditions, exposure to deicing salts and concrete cover permeability and thickness on chloride ingress in concrete. The results show that the chloride ion concentration increases quickly in concrete cover when a structure is exposed to deicing salts at a mountainous location; whereas permeability of concrete cover is an insignificant parameter when the concrete is in direct or splash water contact.

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1. INTRODUCTION

The corrosion initiation period [10] includes the time during which substances such as water, chloride ions and carbon dioxide flow through the concrete cover (i.e. the clear concrete cover protecting the steel reinforcement) and reach the certain concentration necessary to trigger corrosion of the steel reinforcement. The initiation period is characterized by chemical reactions between the various substances and the movement of the various substances in the concrete.

Two models are used, each taking into account a different scale: microscopic and macroscopic models. Microscopic models consider the flow of ions and their chemical balance in the concrete, such as the models Stadium [19], Ms Diff [33], Masi [20], by Shin [31] and Schmidt-Döhl [29][30]. Macroscopic models take into account the various thermal variation as well as hydrous and ionic movements. The chemical reactions are considered only through parameters simulating the chemical effects on transport, like the models by Roelfstra [25], TransChlor [6][8], ClincConc [32], by Meijers [21], Saetta [26] and Ishida [14][18]. While microscopic models simulate the phase changes more precisely and take into account the porosity reduction, they often require extensive testing to obtain valid data for the model parameters.

The TransChlor model is an original model to address chloride ion movement with water in concrete and to consider microclimates [4] [5] reconstituted from real climates. The data is taken from meteorological stations and is reconstituted by considering exposure level of a reinforced concrete structural element. The water movement is accelerated in situations when there adhesion forces between the water and the porous structure of concrete contribute to this movement. This phenomenon is known as capillary suction. The vapour movement is less sensitive to capillarity suction. Instead it follows a diffusion process. Transport of water and water vapour are differentiated in the model [2], [25] [27]. The water transport parameters are obtained from laboratory tests on water adsorption at low temperatures [7] [11].

The TransChlor model basically uses the Fick's diffusion law for water vapour transport [3] [22], thermal diffusion and chloride ion diffusion in water [6]. This law allows the simulation of the diffusion process due to a concentration gradient of these substances. Although the Fick's diffusion law represents vapour diffusion well, it does not describe capillary suction of water as observed in own laboratory tests [6] and by other researchers [15] [25] [28]. Accurate capillary suction modelling is thus obtained by considering the kinetics of the phenomenon.

2. TRANSPORT MODEL

The chloride ion transport in concrete cover is modelled with TransChlor [8]. The relevant parameters considered are the microclimate, the presence of deicing salts and the concrete cover permeability of the structural element. The microclimate is a function of the structural element's exposure to solar radiation (zones in the shade are distinguished from those exposed to the sun), the annual average carbon dioxide concentration and the geographically linked weather conditions (air temperature, relative humidity and precipitation) [9].

The TransChlor model considers the thermal diffusion process and the hydrous transport by capillarity suction and vapour diffusion as a function of the carbonation state, while simulating chloride ion transport in concrete [4] [6] [12]. The various transport processes are modelled with Fick's Equations (1) to (3). The model uses the finite element method to solve for the ion propagation within the concrete and the finite differences method (an implicit method) to solve for the duration of progression of the propagation front.

Equation (1) describes the thermal diffusion process. The specific heat capacity, c_T , depends on moisture content, w, while the thermal conductivity, λ_T , is a function of moisture content, w, and temperature, T [13].

In Equation (2) hydrous transport is modelled by vapour diffusion, D_h , and water capillary, D_{cap} , coefficients. Vapour transport is a function of the temperature, T, and the moisture content, w [3]. The temperature effect is described by means of the Arrhenius law. Water transport is a function of the concrete temperature, T, the moisture content, w, the pore humidity, h_r , and the duration of contact of concrete with water, $t_{contact}$.

The carbon dioxide, CO_2 , transport governs the carbonation progression in concrete. The extent of carbonated concrete is required to determine the amount of chloride ions absorbed by the cement paste. Concrete carbonation depth, x_c , expressed by Equation (3) is obtained by considering the moisture content, the concrete permeability to CO_2 and the chemical carbonation reaction rate [23]. The external conditions are represented by the molar carbon dioxide concentration [CO_2]. The carbonation reaction rate is described by the molar concentrations of calcium hydroxide [$Ca(OH)_2$] and calcium silicate hydrate [CSH]. The concrete carbon dioxide diffusion coefficient, $D_{e,CO2}$, takes into account the concrete permeability and the moisture content evolution in the concrete pores.

$$\frac{\partial T}{\partial t} = \operatorname{div}\left(\frac{\lambda_T(T, w)}{c_T(w)} \cdot \overline{\operatorname{grad}(T)}\right)$$
(1)

$$\frac{\partial h_r}{\partial t} = \operatorname{div}\left(D_h(T, h_r) \cdot \overrightarrow{\operatorname{grad}(h_r)}\right) - \overrightarrow{D_{cap}}(t_{contact}, h_r, E/C, T) \circ \overrightarrow{\operatorname{grad}(h_r)}$$
(2)

$$x_{c} = \sqrt{\frac{2 \cdot [CO_{2}] \cdot D_{e,CO2}}{[Ca(OH)_{2}] + 3 \cdot [CSH]}} \cdot \sqrt{t}$$
(3)

The transport of chloride ions following Equation (4) is a function of the chloride ion diffusion through the pore water defined by moisture content, w, and the movement of entrained chloride ions desolved in water moving through the concrete described by Equation (2) [8]. This phenomenon is referred to as convection. The retardation of the chloride ion front with respect to the convection induced water movement is taken into account by including the retardation coefficient, R_{Cl}. Hardening cement paste and the carbonation rate trap a certain amount of chloride ions [17] [24] [32]. The chloride ions bound in the cement paste, c_b, are distinguished from the free chloride ions moving through the concrete, c_f. The relation between total amount of chloride ions C, c_f and c_b is given by Equation (5), including the Freundlich isotherm with constant parameters β and γ . Chloride ion diffusion coefficient in water, D_{Cl}, varies with the temperature following the Arrhenius law [34].

$$\frac{\partial C}{\partial t} = \operatorname{div}\left(R_{Cl} \cdot c_f \cdot D_h \cdot \overline{\operatorname{grad}(h_r)} + w(h_r, T) \cdot D_{Cl} \cdot \overline{\operatorname{grad}(C)}\right) + R_{Cl} \cdot c_f \cdot \left(\overline{D_{cap}} \circ \overline{\operatorname{grad}(h_r)}\right)$$
(4)

$$C = c_f \cdot w + c_f^{\ \beta} \cdot \gamma \tag{5}$$

3. CHLORIDE ION TRANSPORT BY WATER

The mean velocity or the flow of water is discretised by the finite element method. The positive or negative sign in Equation (6) depends on the direction of water flow.

$$\vec{q} = D_h \cdot \overline{grad(h_r)} \pm \overline{D_{cap}} \cdot h_r \tag{6}$$

 D_{cap} [mm/s] capillarity coefficient

- D_h [mm²/s] water vapour diffusion coefficient
- *H* [-] relative humidity in the concrete pores
- \vec{q} [mm/s] water flow or speed

The weak form can be written as Equation (7).

$$q = \int_{h_L}^{h_R} D_h \cdot [N(x)]^T \cdot [B(x)] \cdot dh \pm D_{cap} \cdot [I] = \frac{D_h}{2} \cdot \begin{bmatrix} -1 & 1 \\ -1 & 1 \end{bmatrix} \cdot \begin{cases} h_1 \\ h_2 \end{cases} \pm D_{cap} \cdot \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \cdot \begin{cases} 1 \\ 1 \end{cases}$$
(7)

[N(x)] interpolation matrix function

[B(x)] matrix of relation for flow and concentration of h

- [*I*] identity matrix
- *x* location in the concrete
- *h* relative humidity at the node

The assembly of all the elements allows calculating the water flow velocity at each node, Equation (8).

$$\begin{cases} q_{0} \\ q_{1} \\ q_{2} \\ \dots \\ q_{n} \end{cases} = \frac{1}{2} \cdot \begin{bmatrix} -D_{h0} & D_{h0} & 0 & \dots & 0 \\ -D_{h0} & D_{h0} - D_{h1} & D_{h1} & \dots & 0 \\ 0 & -D_{h1} & D_{h1} - D_{h2} & \dots & 0 \\ \dots & \dots & \dots & \dots & D_{h,n-1} \\ 0 & 0 & 0 & -D_{h,n-1} & D_{hn} \end{bmatrix} \cdot \begin{bmatrix} h_{0} \\ h_{1} \\ h_{2} \\ \dots \\ h_{n} \end{bmatrix} \pm \begin{bmatrix} D_{cap0} & 0 & \dots & 0 \\ 0 & D_{cap1} & \dots & 0 \\ \dots & \dots & \dots & 0 \\ 0 & 0 & 0 & D_{capn} \end{bmatrix} \cdot \begin{bmatrix} 1 \\ 1 \\ 1 \\ \dots \\ 1 \end{bmatrix}$$
(8)

The average movement of the chloride ions takes into account on each node the delay coefficient and the considered time interval, according to Equation (9).

$$d_{Cl,i} = q_i \cdot R_{Cl} \cdot \Delta t \tag{9}$$

 d_{Cl} [mm] average movement of the chloride ions

 R_{Cl} delay coefficient

$$\Delta t$$
 [s] time interval
i node number

Figure 1a shows that the chloride ion concentration assessment is distorted by using a conventional method [25]. Equation (10) provides the algorithm taking into account chloride ion conservation.



Figure 1: Chloride ion movement: a) inconsistency due to surface superposition, b) transformation on the surface represented by a vector assigned to a node

$$\begin{cases}
B_{0} \\
B_{1} \\
B_{2} \\
\dots \\
B_{n}
\end{cases} = \frac{1}{8} \cdot \begin{bmatrix}
3 \cdot l_{0} & l_{0} & 0 & \dots & 0 \\
l_{0} & 3 \cdot (l_{0} + l_{1}) & l_{1} & \dots & 0 \\
0 & l_{1} & 3 \cdot (l_{1} + l_{2}) & \dots & 0 \\
\dots & \dots & \dots & \dots & l_{n-1} \\
0 & 0 & 0 & l_{n-1} & 3 \cdot l_{n-1}
\end{bmatrix} \cdot \begin{cases}
c_{f0} \\
c_{f1} \\
c_{f2} \\
\dots \\
c_{fn}
\end{cases}$$
(10)

- *B* [kg.mm/m³] surface connecting the chloride ion concentration to the dimension of the finite element
- *l* [mm] finite element length
- c_{fi} [kg/m³] free chloride ion concentration at the ith node, related to the liquid solution

Each surface, B_i , is assigned to the node of displacement vector (Figure 1b) and undergoes a displacement d_i . The surface, B_i , is not inevitably on a node after this displacement. This surface is assigned on the nodes by using the static resolution analogy of a simple beam to take into account the influence of the element distance. As illustrated in Figure 2, the surface, B_i , will be assigned on the node *i*+1 and *i*+2 with Equations (11) and (12) for the example developed in Figure 1.

$$D_{i+1} = \frac{B_i \cdot b}{l_{i+1}} \tag{11}$$

$$D_{i+2} = \frac{B_i \cdot a}{l_{i+1}} \tag{12}$$

D [kg.mm/m³] surface relating the chloride ion concentration to a distance represented by the finite element dimensions

a and b [mm] vector position distances B_i according to the simple beam analogy



Figure 2: Simple beam analogy for assigning surfaces B_i to the node

Finally, the chloride ion concentration is obtained starting from an identical equation to Equation (10), but with the unknown being the vector of the chloride ion concentrations, illustrated as Equation (13).

$$\begin{cases} D_{0} \\ D_{1} \\ D_{2} \\ \dots \\ D_{n} \end{cases} = \frac{1}{8} \cdot \begin{bmatrix} 3 \cdot l_{0} & l_{0} & 0 & \dots & 0 \\ l_{0} & 3 \cdot (l_{0} + l_{1}) & l_{1} & \dots & 0 \\ 0 & l_{1} & 3 \cdot (l_{1} + l_{2}) & \dots & 0 \\ \dots & \dots & \dots & \dots & l_{n-1} \\ 0 & 0 & 0 & l_{n-1} & 3 \cdot l_{n-1} \end{bmatrix} \cdot \begin{bmatrix} c_{f0} \\ c_{f1} \\ c_{f2} \\ \dots \\ c_{fn} \end{bmatrix}$$
(13)

Local equilibrium [1] on the nodes at the extremities is not guaranteed. However, the boundary condition values of the chloride ion concentration in water are assigned to these nodes.

4. VALIDATION

Comparison of measured temperatures of laboratory samples and the TransChlor results validate the thermal transfer simulation model. The sample temperatures are between 20°C and -20°C [6] [7] [8].

The TransChlor model also simulates transfer by means of capillary suction [6] [8]. The model reproduces the chloride ingress due to convection by water well. To validate the modelled water transport, lab test on an immersed sample in brine at low temperatures are performed [7]. The chloride and water front passage are determined experimentally and numerically in a concrete sample with water to cement ratio of 0.52. In the experiment the sample is subjected to the capillary test. A chloride sensor consisting of an optical fibre placed

in the concrete sample at 18 mm from the surface measures the chloride concentration. The results of the measurements and simulations are variable for the first 8 days of the exposure period and become identical thereafter [16].

5. PARAMETRIC STUDY

5.1 Definition of parameters

Deicing salts are spread on roadways in winter periods. As a result, chloride ions come into contact with concrete structural elements through salt-laden mist (water vapour), and the saline water that is either in direct and permanent contact with the elements or splashed on them by passing vehicles.

The relevant variables analysed are the microclimate, the presence and concentration of deicing salts, the properties of the concrete cover and the carbonation depth (Table 1):

- The microclimate depends on the geographical location and takes into account air temperature, air relative humidity and precipitation. The corresponding data have been obtained from three different meteorological stations in Switzerland: the mountainous area in Davos, the Swiss Plateau in Zurich-Kloten and the Lake of Geneva area at Pully.
- The elements are exposed to stagnant water, splash water and mist.
- The concentration of deicing salts evolves in time depending on the application rate and the local weather conditions for a given geographical location. The chloride ion concentration depends primarily on the type of salt spreader, i.e., mechanical spreading (of either solid salt or brine) or automatic spreading (of brine).
- The permeability and thickness of the concrete cover and the corrosion resistance of the reinforcing steel influence the resistance of the structural element. Three different

concrete cover permeabilities are chosen: low permeability Concrete A, average permeability Concrete B and high permeability Concrete C. The effect of concrete cover thickness is investigated for three different values of 15, 30 and 40 mm.

| Parameters | Parameter variation | | |
|--------------------------|---|--|--|
| | | | |
| spreading type | mechanical spreading, automatic spreading | | |
| types of exposure | stagnant water, splash water, mist | | |
| concrete permeability | Concrete A, Concrete B, Concrete C | | |
| geographical location | Zurich-Kloten, Pully, Davos | | |
| concrete cover thickness | 15mm, 30mm, 60mm | | |

 Table 1: Parameters investigated using the TransChlor model

The chloride ingress is much slower under mist exposure than under stagnant or splash water exposure. Therefore, the evolution of chloride ion content in the concrete is simulated over 25 years for elements exposed to stagnant or splash water, and over 50 years for concrete elements subjected to mist exposure.

5.2 Geographical location

Chloride ions migrate rapidly into the concrete cover during precipitation periods with and without the presence of deicing salt. Seasonal variations can significantly influence chloride ion profiles (i.e., chloride ion concentration as a function of concrete depth). Due to the significant chloride ion transport towards larger depth in the concrete, chloride ion profiles may show significant concentrations at the steel reinforcement level during winter periods and reduced concentrations in summer. The space variability of the moisture content develops

differently depending on the type of contact with water (exposure to stagnant or splash water) or water vapour (mist exposure). This has a direct impact on the chloride ion convection by water.

Table 2 shows that the temperature decreases with increasing altitude of the location [6]. The lowest temperatures are recorded in the mountainous area, while the temperatures in the plains are moderate (Figure 3a).

Table 2: Swiss areas with altitudes of each location (A: Altitude, T: Average annual airtemperature, RH: Average annual air relative humidity)

| Areas (location) | A [meters above sea level] | T [°C] | RH [%] |
|-------------------------------|----------------------------|---------------|--------|
| Mountainous (Davos) | 1590 | 2.8 | 73.2 |
| Lake Geneva (Pully) | 461 | 10.4 | 71.7 |
| Swiss Plateau (Zürich-Kloten) | 436 | 8.5 | 77.6 |

The air relative humidity is reflected directly by the relative humidity in the concrete pores for mist exposure. Initial condition considers a completely saturated concrete. The Swiss Plateau is the wettest climate, followed by the mountainous and the Lake of Geneva areas, respectively (Figure 3b). The relative humidity in the concrete pores is directly related to the precipitations in a given area and class of exposure as shown in particular for splash exposure. Figure 3c shows the fluctuations and the high values of relative humidity in concrete pores for the mountainous area due to the significant precipitations in the region.

In Figures 3d and 3e, the simulation results for splash and mist exposure zones illustrate that for low moisture level the rate of concrete carbonation is faster. As shown in the figures, the rate of carbonation is slowest for the Swiss Plateau, where the air relative humidity is

higher than for the Lake of Geneva and mountainous areas for which the rate of carbonation of concrete are similar.





Figure 3 : Influence of geographical location on chloride ion transport as simulated with TransChlor

Under mist exposure, chloride ion ingress is similar for all three areas (Figure 3f). This evolution is explained by the similar relative humidity level in each location. The chloride ion diffusion becomes important and is similar for each location.

The evolution of chloride ion ingress under splash exposure is fastest in mountainous areas (Figure 3g). For the Swiss Plateau, this evolution is slow in the initial stage, but it rates increases. In about 10 years it reaches values close to the ones for the mountainous area, and after it remains constant. The chloride ion concentration evolution remains rather slow for the Lake of Geneva area because the amount of deicing salt spread on the road is less than the other areas.

5.3 Exposure of the structural element

The presence of water on a structural element is taken into account by stagnant water and splash exposure. The stagnant water and splash exposure zones have a similar evolution of relative humidity while mist exposure has a smaller fluctuation in relative humidity (Figure 4a). The relative humidity under mist exposure remains constant at about 83% after 10 years, which is about 10% higher than the average annual air relative humidity.



Figure 4: Influence of structural element exposure as simulated with TransChlor on a) relative humidity of concrete and b) total chloride content

Chloride ion ingress into concrete cover is much more significant for the stagnant water and splash exposures than for mist exposure (Figure 4b). Furthermore, the chloride ion evolutions for former two cases are similar. The contact time of the structure with water is not different for these two exposures. This contact time comprises a large variability for the stagnant water exposure and is directly related to the drying time of water trapped in the concrete near the surface.

Figure 5a demonstrates that a significantly larger amount of chloride ions migrate into concrete under splash exposure than under mist exposure. Maximum chloride ion concentration occurs at a depth away from the surface. The concentration of chloride ions at the concrete surface is rather low. This is due to the transport of the ions by the capillary

suction. Figure 5b shows that the chloride profile after 5 years is similar to the one after 25 years; thus, the maximum chloride ion concentration is reached after 5 years. With time chloride ions only migrate further into the concrete but chloride ion concentration does not increase.



Figure 5: Spatial variation of the total chloride content in concrete for different a) type of exposure and b) time of exposure

5.4 Type of deicing salt spreading

The amount of deicing salt spread on the roadway depends on the chosen spreading type, i.e., the mechanical or automatic spreading. Typical amounts of salt spread in a mountainous area for mechanical and automatic spreading are 1497 and 1213 [g/m².year], respectively [6].

The chloride ion evolution in concrete is slower in the case of automatic spreading (Figure 6). For mist exposure after fifty years, the chloride ion concentration amounts to approximately 45% of that calculated for mechanical spreading.



Figure 6: Influence of spreading type on chloride ion concentration for a) mist and b) splash exposure

5.5 Concrete permeability

Movements of substances in concrete depend directly on the concrete permeability for all transport modes, except for the thermal transfer. Figure 7 shows that the ingress of chloride ions, propagation of carbonation and evolution of relative humidity are most significant for Concrete C with the highest permeability.

The pores filling rate is illustrated for two different types of exposure: splash water and mist exposure. The wetting and drying fluctuations are faster in the Concrete C for zones expose to splash water (Figures 7b and 7c).



Figure 7: Influence of concrete permeability on the various transport modes as simulated with TransChlor

In comparison to Concrete C, the carbonation depth is reduced to about one third for Concrete A and to one half for Concrete B (Figure 7a). The results in Figure 7 also indicate that there is only a small influence of permeability on the chloride ion concentration. In fact, after 10 years of splash exposure, the chloride content is the same for all three types of concretes.

The results of chloride concentrations from the simulations are frequently observed in structural elements exposed to deicing salts due to splash exposure for more than 20 years, when they show significant signs of corrosion requiring rehabilitation.

5.6 Concrete cover thickness

The concrete temperature changes very quickly with the external environment. Therefore, instead of a thermal diffusion model for predicting the change of the temperature in the concrete cover, the meteorological temperature data with a time step of more than one hour can be used (Figure 8a). The hydrous transfer attenuates the relative humidity gradients in the concrete depth. This attenuation is more important for thicker members under mist exposure (Figures 8b and 8c). The chloride ion evolution in the concrete cover is particularly important in the case of splash exposure. Depths of more than 60 millimeters show an already important attenuation of the chloride concentrations at 25 years. The rate of chloride ion evolution in the concrete, however, remains slow for a salt-laden mist exposure (Figures 8d and 8e).



Figure 8: Influence of concrete thickness on the various transport modes as simulated

with TransChlor

6. CONCLUSIONS

Water movement by capillary suction is modelled with kinematic equations. This movement is transformed into chloride ion movement due to the chloride ion convection in water. This phenomena is implemented in the TransChlor Model to simulate chloride ingress into concrete cover. The model also considers reconstituted microclimatic conditions. The following conclusions can be drawn from the TransChlor numerical simulations:

- 1. Chloride ion concentration increases quickly in the concrete cover when structural elements are exposed to deicing salts and water (splash and direct contact).
- Maximum chloride content is reached after less than 10 years at a depth of about 20 to 40mm from the surface.
- 3. Chloride ingress significantly depends on the climatic condition and the amount of deicing salts applied. Permeability of concrete cover is thus a rather insignificant parameter when the concrete is in direct and splash water contact.

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