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# Modelling of chloride migration in cementitious materials including thermodynamic equilibria

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## Résumé

*Dans cet article, une approche multi-espèces pour la modélisation du transfert réactif d'ions chlorure dans les matériaux cimentaires est proposée. Elle considère les équilibres thermodynamiques dans le matériau qui traduisent les interactions liquide – solide et les phénomènes de dissolution/précipitation lors du transfert : dissolution et précipitation de Portlandite et de C-S-H. Le flux des ions considérés est calculé en utilisant l'équation de Nernst-Planck qui intègre la diffusion naturelle sous un gradient de concentration, la migration sous un champ électrique et l'activité ionique. Le modèle proposé est appliqué pour simuler l'essai de migration de chlorures en régime stationnaire sur deux pâtes de ciment. La première est composée uniquement du ciment Portland CEM I 52.5 N, tandis que la deuxième pâte contient des cendres volantes. Les résultats numériques des flux de  $\text{Cl}^-$ ,  $\text{Na}^+$  et  $\text{K}^+$  en régime stationnaire ont permis de calculer les coefficients de diffusion effectifs de ces ions.*

## Abstract

*In this paper, a multispecies approach for the reactive transport modelling in cementitious materials is proposed. It considers the thermodynamic equilibria in the material during the transport that reflect the liquid – solid interactions and the dissolution/precipitation of solid phases (minerals): dissolution and precipitation of Portlandite and C-S-H. The ion fluxes during the transport are calculated using the Nernst-Planck equation that considers the natural diffusion under a concentration gradient, the migration under an electrical field, and the chemical activity. The model proposed is applied to simulate the standard chloride migration test on two cement pastes. The first paste is based only on Portland cement, while the second paste is composed of fly ash. Numerical results of fluxes of  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{K}^+$  at the steady state are used to calculate the corresponding effective ion diffusion coefficients in the material studied.*

**Mots clefs :** Durabilité ; Modélisation ; Essai de migration ; Matériaux cimentaires ; Coefficient de diffusion des chlorures.

**Keywords :** Durability ; Modelling ; Migration test ; Cementitious materials ; Chloride diffusion coefficient.

## 1. Introduction

Reinforced concrete (RC) structures in marine areas are affected by chloride attack. Chlorides penetrate the cover concrete, through the porosity, under a difference in concentration and/or liquid pressure (convection). The chlorides reach the rebars and, when they reach a threshold concentration, they generate their corrosion that can cause local or general degradations of the structures, i.e. reducing the structure service life. Given the high costs of the maintenance/repair, the durability of RC structures in their environments is nowadays one of the main challenges mentioned in the specifications of large construction projects. The development of more accurate models of reactive transport in concrete will improve the prediction of the structure service life.

In the last decades, several experimental and numerical studies were developed in the literature to propose methods and tools for predicting chloride transport in cementitious materials. We quote the standard migration tests used to determine the chloride diffusion coefficient in the steady state or non-steady state [1]–[7]. Also, several approaches for chloride transport modelling were developed in order to predict chloride ingress in concrete with limited resources and time [8]–[15]. Amiri et al. [16] were among the first researchers to develop a single-species approach for chloride transport modelling, considering diffusion and migration phenomena. After that, many multispecies approaches were developed, considering several ions in the pore solution and more chemical and physical interactions during the transport (chemical activity and electrical double layer “EDL” [9], [13], [17]–[21]).

Moreover, recent researches highlight the impact of the thermodynamic equilibria on the chloride reactive transport in cementitious materials [22]–[26]. Tran et al. [27], [28] proposed a coupling between chloride transport and thermodynamic equilibria by adding kinetic control for predicting chloride binding in concrete. Jensen et al. [29], [30] developed a multispecies approach for the reactive mass transport through cementitious materials considering chemical equilibria by using geochemical code PhreeqC<sup>®</sup>. The modelling was applied on saturated mortars exposed to NaCl and seawater for 180 days. Yu et Zhang [31] developed a new approach to simulate leaching of cement paste in ammonium nitrate solution considering mass transport, chemical kinetics and thermodynamic equilibria. Poisson-Nernst-Planck equation was used to simulate the mass transport, while the chemical activity was calculated using Davies’ model. Cherif et al. [32] proposed a physico-chemical modelling of multispecies transport throughout cementitious materials considering thermodynamic equilibria (Portlandite dissolution and Friedel’s and Kugel’s salts precipitation). Ion fluxes were calculated using the Nernst-Planck equation.

The studies mentioned above highlight the need to include thermodynamic equilibria in the modelling of chloride transport in cementitious materials. However, the models are based on synthetic pore solutions with a limited number of ions and solid phases. Also, some models are based on Fick’s law that misdescribes the mass transport in concrete without considering ionic interactions.

In this paper, a multispecies modelling coupling reactive transport and thermodynamic equilibria was proposed. Nernst-Planck equation was used to simulate the mass transport, while the thermodynamic equilibria and the kinetics of dissolution/precipitation were calculated by using the geochemical code PhreeqC<sup>®</sup>. They reflect the dissolution and precipitation of Portlandite and C-S-H during chloride transport in cementitious materials. The model proposed is applied to simulate the chloride migration over two cement pastes under an applied electrical field, i.e. to simulate the standard migration test. The first paste is based only on Ordinary Portland, while the second paste is composed of fly ash. The migration cell is composed of a basic solution in the upstream and downstream compartments: 25 mM of NaOH and 83 mM of KOH. In addition, 500 mM of NaCl is

added on the upstream (boundary conditions). The real composition of the pore solution of the paste tested is considered as initial conditions. The model outputs are as follows:

- (i) The ion profiles in the material that can be used for the calculation of the ion diffusion coefficients in the non-steady state (from the ion penetration depth);
- (ii) The ion fluxes in the downstream that can be used for the calculation of the ion diffusion coefficients in the steady state;

Finally, the effective chloride diffusion coefficient obtained from the modelling proposed was compared to that obtained from flux measurements at the steady state.

## 2. Methodology

### 2.1. Modelling principle

The time evolution of the ion  $i$  concentration ( $C_i$ ) in the migration cell is calculated by the mass balance equation (Eq.(1) which considers the ion transport (ion fluxes  $J_i$ ) and the interactions between ions and the material tested ( $q_i$ ). Ion fluxes are calculated using Nernst-Planck equation considering the diffusion, migration and chemical activity (see Eq. (2). Noted that the proposed modelling focuses on ion transport in saturated materials that does not require the coupling with convection and heat and humidity transfer.

$$\frac{\partial C_i}{\partial t} = -\nabla J_i - \frac{\partial q_i}{\partial t} \quad (1)$$

where,

$$J_i = -D_{E,i} \left( \nabla C_i + C_i \nabla \ln \gamma_i + \frac{z_i C_i F}{RT} \nabla \psi \right) \quad (2)$$

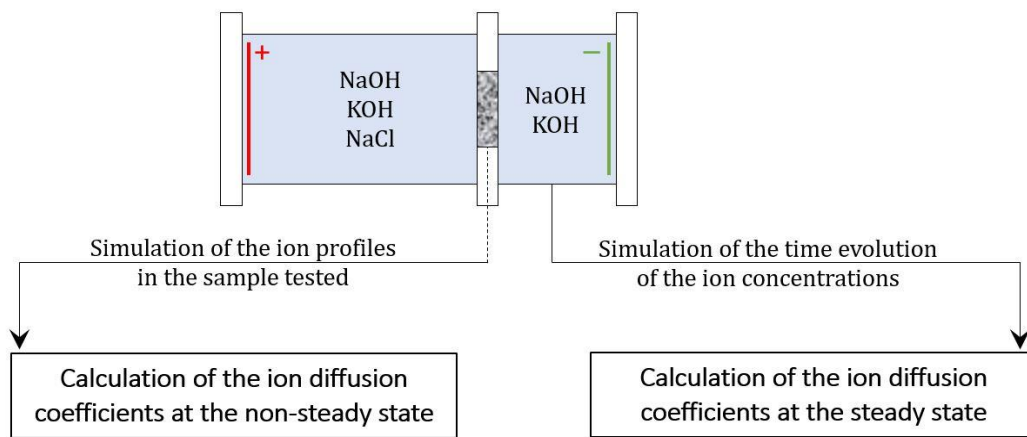
where  $D_{E,i}$  [ $\text{m}^2 \cdot \text{s}^{-1}$ ] is the effective diffusion coefficient,  $\gamma_i$  is the activity of the ion  $i$ ,  $\psi$  [V] is the total electrical potential,  $z_i$  is the valence of ion  $i$ ,  $F$  [ $\text{C} \cdot \text{mol}^{-1}$ ] is the Faraday constant,  $R$  [ $\text{J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$ ] is the ideal gas constant,  $T$  [K] is the temperature,

The term “ $q_i$ ” is calculated from a pure thermodynamic approach using kinetic laws and the thermodynamic equilibrium constants ( $K_s$ ) of the considered solid phases of the material tested [25]–[28]. It considers the dissolution / precipitation of the solid phases during the migration. The solid phases considered for this study are: C-S-H; Portlandite, Friedel’s and Kugel’s salts precipitated in pores during the chloride migration, while the ions considered are:  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$ . The resolution of the mathematical system was done using the geochemical code PhreeqC<sup>®</sup>. The command [TRANSPORT] was used to calculate the ion fluxes, while the commande [EQUILIBRIUM PHASES] was used to simulate the thermodynamic equilibria.

### 2.2. Case study

The multispecies approach proposed allows simulating all the ion movements in the migration cell during the standard migration test. In this study, we simulate the time evolution of ion concentration in the downstream compartment and ion concentration profiles in the material tested, considering the dissolution and precipitation of C-S-H, portlandite, Friedel’s and Kugel’s salts in the pores (see

Fig. 1). The simulation of the ion concentrations in the downstream allows calculating the corresponding ion fluxes and the diffusion coefficients in the steady state [5], [33], while the ion profiles in the material are useful for the calculation of the diffusion coefficient at the non-steady state [3], [34]. A standard migration test based on basic solution in the upstream and downstream compartments was performed: 25 mM of NaOH and 83 mM of KOH (boundary conditions). Moreover, 500 mM of NaCl is added on the upstream. An electrical field of  $300 \text{ V.m}^{-1}$  was applied at the sample sides and monitored using two calomel reference electrodes at the two sides of the material tested (sample of 6.3 cm diameter and 1 cm thickness).



**Fig. 1.** Schematic representation of the modelling procedure performed for this study.

Finally, the modelling is applied on two cement pastes submitted to the standard migration test during 5 days: (i) paste 1 with only Ordinary Portland Cement and a water to cement ratio (W/C) of 0.5; (ii) paste 2 with Ordinary Portland Cement and fly ash (mass substitution of 30% of cement) and water to binder ratio (W/B) of 0.4. The chemical composition of the pore solution of paste 1 and paste 2 are taken from the literature (initial conditions) [7], [35]. Initial and boundary conditions of the pastes 1 and 2 are given in Table 1.

**Table 1.** Initial and boundary conditions for pastes 1 and 2.

	Boundary conditions		Initial conditions	
	Upstream ( $x=0,t$ ) [mol.m <sup>-3</sup> ]	Downstream ( $x = L, t$ ) [mol.m <sup>-3</sup> ]	(Paste 1) ( $t=0, 0<x<L$ ) [mol. m <sup>-3</sup> ] [7], [35]	(Paste 2) ( $t=0, 0<x<L$ ) [mol. m <sup>-3</sup> ] [7], [35]
$C_{Cl^-}$	500	0	4	1
$C_{Na^+}$	525	25	51	48
$C_{K^+}$	83	83	117	50
$C_{Ca^{2+}}$	0	0	2	2
$C_{SO_4^{2-}}$	0	0	2	2

### 3. Results and discussion

#### 3.1. Ion profiles in the material tested

Figs. 2 and 3 give the simulated profiles of  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  in the pastes 1 and 2 at the end of the migration test (after 5 days). The profiles simulated highlight a high chloride concentration in the sample side exposed to NaCl for the two pastes studied. Chlorides diffuse from the upstream to the sample tested under the applied electrical field. However, it is necessary to remark the surprising event that the depth of the maximum chloride concentration is only of 5 mm and 7 mm for the sample pastes 1 and 2, respectively (chloride front) in spite the steady state seems to be reached (as discussed in the following section). The chloride concentration in the rest of the depth is low, whereas it should be maximum along the sample depth according to the literature [1], [16], [36]. At this respect, it has to be mentioned the possibility that the chloride decrease in the rest of the depth is forced by the electroneutrality of the pore solution since the  $\text{OH}^-$  and the electrode reactions, which ensure the electroneutrality, are neglected. Moreover, we noted an increase of the concentrations of  $\text{Na}^+$  and  $\text{K}^+$  in the pore solution due to their migration from the downstream to the sample tested. Finally, the increase of  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  is due to the dissolution of portlandite and monosulfoaluminates under the electrical field [37], [38].

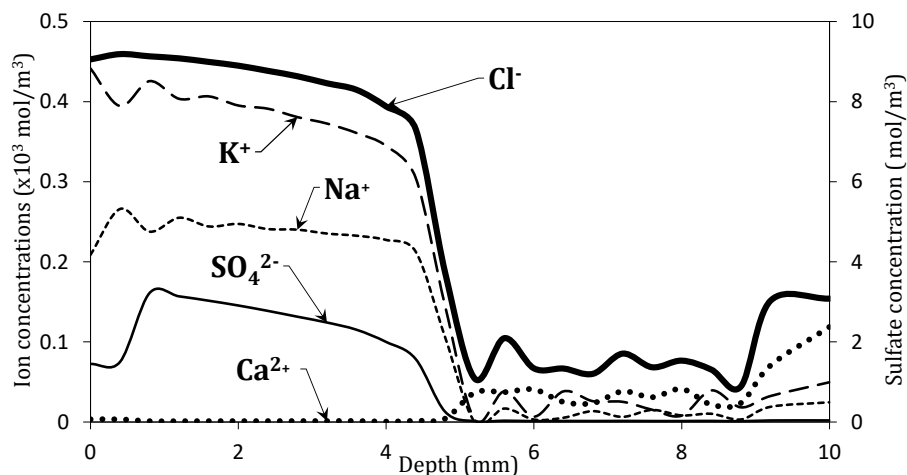


Fig. 2. Profiles of  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$  in the pastes 1 at the end of the migration test (5 days).

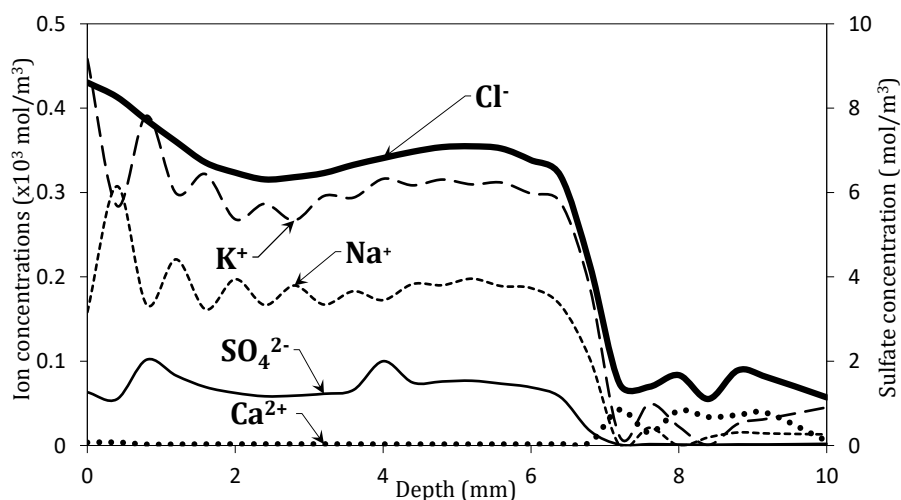
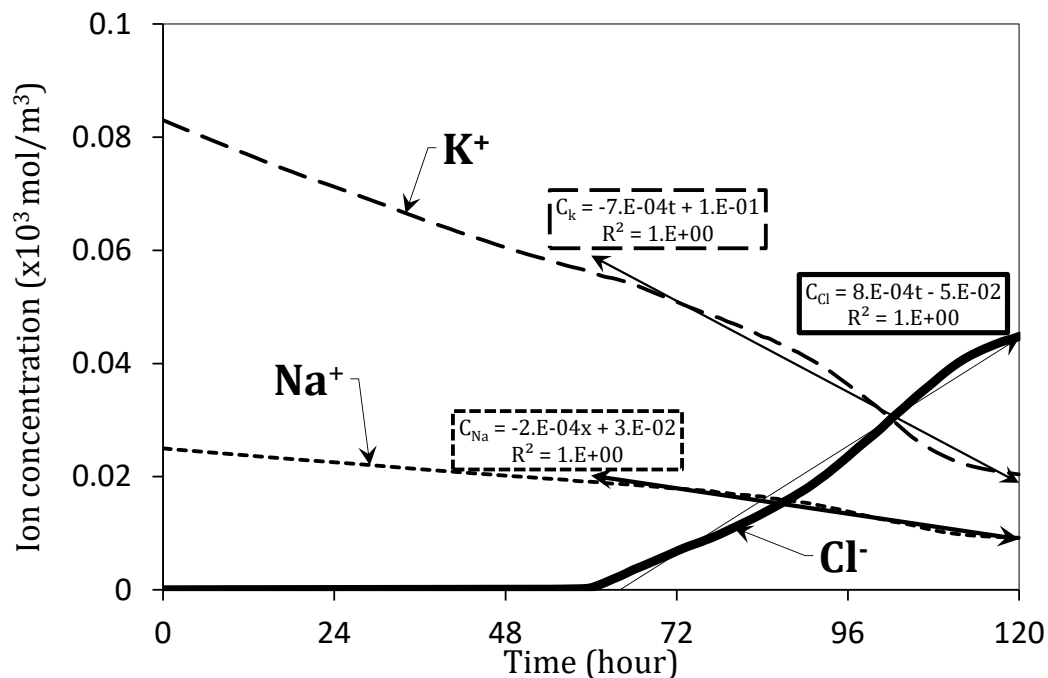


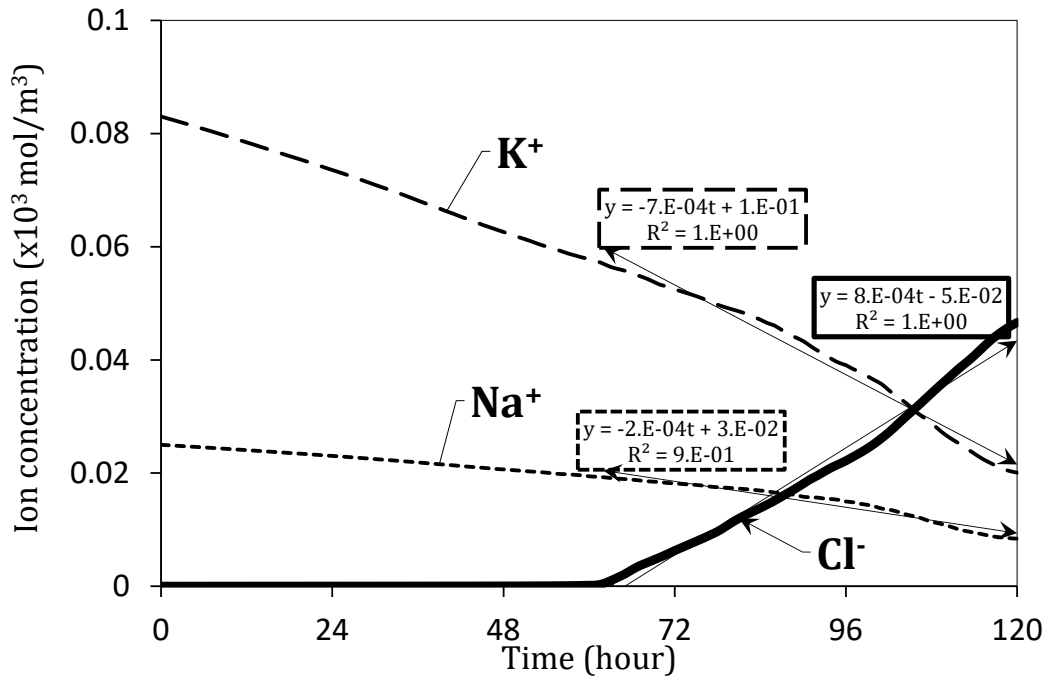
Fig. 3. Profiles of  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$  in the pastes 2 at the end of the migration test (5 days).

### 3.2 Ion fluxes in the downstream

Figs. 4 and 5 show the time evolutions of concentration of  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{K}^+$  in the downstream.  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  concentrations in the downstream being negligible, they are not illustrated. The chloride gain is due to their migration from the upstream under the applied electrical field and possible diffusion under the concentration gradient between the two compartments considering the short time of the migration test. During the first 60 hr-migration test, chloride concentration in the downstream was zero, i.e. chloride has not yet reach the downstream. It corresponds to the “time lag.” It makes chloride to pass through the sample tested (about 60 hr). After that, we note that the steady state is reached with a chloride migration kinetic of  $8\text{E-}3 \text{ mol.m}^{-3}.\text{s}^{-1}$  for the pastes 1 and 2. The chloride migration kinetics of the two pastes is identical since the pore solution compositions of the two pastes are relatively similar. Moreover, the loss in sodium and potassium in the downstream is due to their migration to the upstream (with same kinetics of the two pastes) under the applied electrical field. Their migration kinetics from the downstream to the material and the upstream are of  $2\text{E-}1 \text{ mol.m}^{-3}.\text{s}^{-1}$  and  $7\text{E-}1 \text{ mol.m}^{-3}.\text{s}^{-1}$  for  $\text{Na}^+$  and  $\text{K}^+$ , respectively.



**Fig. 4.** Time evolution of the concentration of  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{K}^+$  in the downstream of paste 1.



**Fig. 5.** Time evolution of the concentration of  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{K}^+$  in the downstream of paste 2.

The fluxes of  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{K}^+$  in the downstream are calculated from the time evolution of the corresponding concentrations (kinetics). The fluxes are given in Table 2. *Ion fluxes in the downstream for the pastes tested*

**Table 2.** Ion fluxes in the downstream for the pastes tested.

	$\text{Cl}^-$	$\text{Na}^+$	$\text{K}^+$
Paste 1 [ $\times 10^{-5} \text{mol.m}^{-2}.\text{s}^{-1}$ ]	7.1	-1.8	-6.2
Paste 2 [ $\times 10^{-5} \text{mol.m}^{-2}.\text{s}^{-1}$ ]	7.1	-1.8	-6.2

### 3.3 The effective coefficient diffusion

Firstly, the chloride diffusion coefficient in the pore solution is calculated from the chloride flux given in Table 2 using Eq. (3) [16].

$$D_{\text{Cl,PS}} = \frac{RTL}{Z_{\text{Cl}} - FU} \frac{J_{\text{Cl}}}{C_{\text{Cl}}} \left( 1 - e^{\frac{z_{\text{Cl}}^- FU}{RT}} \right) \quad (3)$$

where  $L$  [m] is the thickness of the sample;  $C_{\text{Cl}}$  [ $\text{mol.m}^{-3}$ ] the chloride concentration in the upstream;  $U$  [V] the potential difference at the sample terminals.

Next, the effective chloride diffusion coefficients of the both pastes tested are calculated from the corresponding chloride coefficient diffusion in the pore solution and the formation factor  $f$  (see Eq. (4)). For this study, we consider that the formation factor depends only on the porosity “ $\varphi$ ” according to, [27], [28]. The effective diffusion coefficients calculated and those experimentally measured are given in Table 3. The diffusion coefficients calculated and measured are relatively similar with ratios of about 3:4 and 3:2 for pastes 1 and 2, respectively. The differences in the diffusion coefficients seem to be due to errors related to the chemical interactions, neglected in the modelling (EDL, formation of



monosulfoaluminates) and the variation of the microstructure of the pastes during the chloride migration [37], [39].

$$D_{Cl,eff} = f \times D_{Cl,PS} = \varphi \times D_{Cl,PS} \quad (4)$$

**Table 3.** Chloride diffusion coefficients of the pastes tested.

	Paste 1	Paste 2
$\varphi$ [%] [7]	34.2	29.4
$D_{eff}$ [ $\times 10^{-12} \text{m}^2 \cdot \text{s}^{-1}$ ] « experimentally measured »	5.6	2.4
$D_{eff}$ [ $\times 10^{-12} \text{m}^2 \cdot \text{s}^{-1}$ ] « calculated »	4.1	3.6

## 4. Conclusion

From the obtained results, following main conclusions are made:

- The modelling proposed allows simulating the standard migration test at the steady and non-steady states considering the real pore solution of the material tested and the dissolution/precipitation phenomena during the migration. The modelling was applied on low clinker-cement pastes based on Portland cement and fly ash.
- The output of the modelling proposed are the ion profiles in the material tested and time evolution of the ion concentrations in the upstream and downstream compartments during the migration test. The ion profiles are useful for the calculation of the ion diffusion coefficients in the non-steady state, while the concentration evolutions in the downstream are useful for the calculation of the ion diffusion coefficients at the steady state.
- The concentration evolutions in the downstream of  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{K}^+$  were used to calculate the fluxes and diffusion coefficients of these ions for the two pastes tested. Next, the fluxes and diffusion coefficients calculated are compared to those experimentally measured. Promising results are highlighted which would allow simplifying the calculation of chloride diffusion coefficients of cementitious materials from modelling. The model proposed could be improved considering more solid phases such as, oxychloride, ettringites and electrode reactions during the migration test.

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