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Mohammed Naji HAMMOOD

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A Meso-Macro Numerical Approach for Chloride Diffusivity Modeling Taking into Account Chloride Binding and Crack Evolution in Concrete

		JURY
Président du jury	Abdelkarim AÏT-MOKHTAR	Professeur, Université de La Rochelle
Rapporteurs :	Jean Baptiste COLLIAT	Professeur, Université de Lille 1
	Fabrice BERNARD	Maitre de Conférences - HDR, INSA de Rennes
Examinateurs :	Abdelkarim AÏT-MOKHTAR	Professeur, Université de La Rochelle
Invitée :	Stéphanie BONNET	Maitre de Conférences, Université de Nantes
Directeur de Thèse :	Ouali AMIRI	Professeur, Université de Nantes
Co-directeur de Thèse :	Nathan BENKEMOUN	Maitre de Conférences, Université de Nantes

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Abstract

The penetration of chloride ions has an essential responsibility in the degradation of concrete structures caused by reinforcement corrosion leading to a severe impact on the durability and service life of concrete structures. The problem becomes more critical with the existence of cracking which accelerate the penetration of chloride ions into concrete cover. In this work, the FE formulation for the numerical modelling of chloride ions diffusion accounting for chloride binding capacity in mesoscale concrete is introduced. The mesostructure is based on a two-phase 3D representation of heterogeneous materials, such as concrete, where stiff aggregates are embedded into a mortar matrix. For this purpose, we turn to the Embedded Finite Element Method (E-FEM). This is performed by introducing a weak discontinuity in the chloride concentration field for finite elements where the physical interface is present. Numerical spatial homogenization experiments based on work Pouya are also performed on 3D mesostructures to compute macroscopic diffusivity tensors accounting for two-phase material. Comparison with Maxwell's equation and experimental results are carried out to show the accuracy of the proposed numerical approach. Finally, the meso-macro approach is presented to introduce a numerical model capable of providing macroscopic information (mean diffusivity tensor) integrating the level of crack opening, crack path and heterogeneity of materials in quasi-brittle concrete. The mesoscale coupling with the mass transport part is based on Fick's Law with a modified diffusion coefficient taking into account crack opening and aggregates. The macroscopic diffusivity tensor integrates more complex features such as the cracking evolution process, tortuosity of the crack's path, induced-anisotropy and presence of aggregates. The defined tensor is used afterwards in order to estimate the service-life of concrete structures, including the effect of the cracking and the internal mesostructure.

Résumé

La pénétration des ions chlorure est la principale cause de la dégradation des structures en béton, par corrosion des armatures, entraînant un impact sévère sur leur durabilité et leur durée de vie. La pénétration de ces agents agressifs pourrait être favorisée davantage par la présence des fissures. Dans cet thèse, nous avons utilisé la méthode des éléments finis (EF) pour résoudre l'équation de la loi de Fick couplée à la capacité de fixation d'ions chlorure afin de modiliser la diffusion des ions chlorure à l'echelle mésoscopique. Dans un premier temps, nous avons considéré une représentation 3D d'un matériau, sain, hétérogène biphasé (comme le béton) ou les inclusions (granulats) sont noyées dans une matrice de mortier. Le problème des interfaces (inclusion/matrice) a été résolu en utilisant la méthode E-FEM (Embedded Finit Element Method). Au niveau de ces interface, nous avons introduit une discontinuité faible du champ de concentration de chlorure. Une approche d'homogénéisation par moyennes spatiales se basent sur les travaux de Pouya est également utilisée pour prédire les tenseurs de diffusivité macroscopiques des matériaux biphasiques. La comparaison avec l'équation de Maxwell et des résultats expérimentaux a été réalisée pour montrer la précision de l'approche numérique proposé.

Dans un second temps, l'approche méso-macro est représentée pour introduire un modèle numérique capable de fournir des informations macroscopiques (tenseur de diffusion moyen) intégrant le niveau d'ouverture de fissure, le chemin de fissuration et l'hétérogénéité d'un matériau quasi fragile tels que le béton. Dans ce cas, des points clés du processus de fissuration comme l'évolution d'une fissuration répartie dans l'éprouvette vers une fissuration localisée (macro-fissure(s)), comme la tortuosité de la fissure et l'anisotropie de la fissuration sont intégrés naturellement dans la diffusivité macroscopique. En fin, le tenseur défini est ensuite utilisé afin d'estimer la durée de vie des structures en béton, y compris l'effet de l'endommagement et de la méso-structure interne.

Résumé Étendu

Contexte général et étude bibliographique

Dans le domaine du Génie Civil, mettre en place une méthodologie de développement durable intégrant la pérennité des constructions ou infrastructures est aujourd'hui primordial. Cette pérennité passe par (1) une meilleure compréhension du comportement à long terme des matériaux cimentaires sous divers sollicitations environnementales agressives et (2) par l'évaluation des mécanismes de dégradation alors associés (vieillissement, perte de capacité portante et risque de rupture). Dans le cadre des infrastructures du Génie Civil en béton armé, des agents agressifs provenant d'un milieu extérieur tels que l'eau de mer et les embruns marins et/ou les sels de déverglaçage, ou des matériaux entrant eux-mêmes dans la composition d'un béton (sable ou certains adjuvants), sont susceptibles de pénétrer dans le matériau cimentaire. Les agents les plus critiques vis-à-vis du béton armé sont les ions chlorure. En pénétrant peu à peu dans le béton d'enrobage, ils peuvent atteindre les armatures, détruire la couche passive qui les protège et corroder les armatures, affectant ainsi la durabilité de l'infrastructure par perte de ses performances mécaniques [Mehta and Monteiro, 1993. Par conséquent, la durabilité et la durée de vie de la structure sont impactées. Le taux de pénétration des ions chlorure dépend fortement des propriétés microstructurales et des composants du béton. Par ailleurs, la fissuration peut accélérer considérablement cette pénétration d'agents agressifs au sein du béton. Elle fournit des passages d'écoulement préférentiel et favorise ainsi la pénétration d'ions chlorure dans le béton. Par conséquent la modélisation de la fissuration tenant compte explicitement des différents constituants du béton revêt un aspect important quant à une prédiction réaliste de la durée de vie des ouvrages. En ce qui concerne les différents constituants du béton, plusieurs échelles (macroscopique, mésoscopique, microscopique et nanoscopique) peuvent être considérées pour analyser le comportement du béton. En fonction de la puissance de calcul, des connaissances mécaniques, chimiques, physiques et numériques, une communication entre ces différentes échelles permet de modéliser avec précision le comportement du béton. À l'échelle macroscopique (l'échelle de la structure), le matériau est considéré comme homogène et le comportement est caractérisé au travers de lois phénoménologiques telles que la plasticité et l'endommagement [Lemaitre et Chaboche, 2004]. Néanmoins, ces modèles phénoménologiques ne prennent pas en considération l'impact de la microstructure et des propriétés de chaque constituant individuel du béton. Il est difficile de décrire correctement les mécanismes physiques (rupture, dégradation ou mécanismes de transport) prenant naissance à une échelle plus fine et responsables du comportement macroscopique du béton. Notons aussi qu'à l'échelle nano et microscopique, l'existence de différentes phases liées aux différents constituants du béton, induisent une complexité dans la modélisation des phases et leur comportement. Cette complexité rend les coûts des calculs assez prohibitifs. À l'échelle mésoscopique, les hétérogénéités sont moins marquées ; le béton peut être considéré comme un matériau bi-phasique : agrégats inclus dans une matrice de mortier. Un modèle à cette échelle est considéré dans cette thèse pour caractériser le matériau et fournir une représentation explicite de l'hétérogénéité (forme, distribution, taille et comportement) présente à l'échelle fine et jouant un rôle fondamental dans l'observation de nombreux phénomènes physiques à l'échelle macroscopique avec un temps de calcul assez raisonnable. Les propriétés de transfert sont influencées par de nombreux paramètres d'interaction liés aux compositions difficilement identifiables par des essais expérimentaux au laboratoire [Abyaneh et al., 2013] d'où l'utilité de prévoir ces propriétés de transfert en fonction de la mésostructure du béton par des modèles analytiques ou numériques. Au cours des 10 dernières

années, la simulation du transfert d'agents agressifs au sein des matériaux cimentaires à l'aide de modèles définies à l'échelle mésoscopique a connu un essor. Ces modèles sont apparus efficaces dans la modélisation des caractéristiques principales du comportement du béton, non seulement dans les aspects mécaniques [Benkemoun et al., 2010], [Roubin et al., 2015] et [Comby Peyrot et al., 2009], mais aussi dans l'aspect transfert de masse (Jourdain et al. [Jourdain et al., 2014b], Benkemoun et al. [Benkemoun et al., 2015] et Grassl et Bolander [Grassl and Bolander, 2016]). Par conséquent cette approche mésoscopique a été retenue dans cette thèse. Concernant le problème de diffusion, Garboczi et Bentz [Garboczi and Bentz, 1998] ont proposé un modèle multi-échelle pour la prédiction de la diffusivité du béton en tenant compte de l'effet de l'auréole ou de l'interface de transition (ITZ). Zheng et al. [Zheng et al., 2012] ont travaillé sur des modèles treillis pour évaluer la diffusion de chlorures dans le béton en simulant sa mésostructure. De plus, l'effet de la forme des agrégats sur la diffusivité d'ions chlorure a été examinée. Zeng [Zeng, 2007] a utilisé la méthode des éléments finis avec un modèle de structure bi-dimensionnelle pour analyser le comportement de la diffusivité de chlorures dans le béton. Il a considéré le béton comme un matériau bi-phasique constitué d'agrégats inclus dans un mortier. Li et al. [Li et al., 2012] ont également utilisé la méthode des éléments finis pour prédire la diffusivité d'ions chlorure dans le béton avec un modèle à deux et trois dimensions tenant compte de la forme bi-phasique du béton. L'effet de l'ITZ a été pris en considération dans les travaux d'Abyaneh et al. [Abyaneh et al., 2013] pour l'estimation du coefficient de diffusion de chlorures à l'échelle mésoscopique. Wang et Ueda [Wang et Ueda, 2009] ont utilisé un modèle de réseau de treillis à deux dimensions à l'échelle mésoscopique pour estimer la diffusion de chlorures dans le béton en fonction du temps et de la température. Notons aussi que le transport d'ions chlorure dans le béton n'est pas un simple processus de diffusion. Il est souvent associé à de différents phénomènes tels que la migration (interaction électrostatique) et l'interaction de chlorures avec la matrice cimentaire physiquement et chimiquement. De nombreuses recherches ont été menées concernant l'étude de la diffusivité d'ions chlorure dans les matériaux cimentaires sous l'effet de différents mécanismes de transport (perméabilité, migration, fixation du chlorure) [Wang and Ueda, 2009, Wang and Ueda, 2011b, Krabbenhøft and Krabbenhøft, 2008, Zeng, 2007. D'autre part, la modélisation du transport des chlorures est représentative de la réalité uniquement si elle intègre le mécanisme de dégradation principal des structures en béton en service, à savoir la fissuration. En effet, cette dernière est responsable, tout comme les pores du matériau, d'une pénétration des ions chlorures mais cette pénétration devient privilégiée [Djerbi et al., 2008] et provoque un vieillissement accéléré de l'infrastructure concernée dès lors que la fissuration localisée (présence d'une macrofissure par exemple) prend le pas sur la fissuration diffuse [Djerbi-Tegguer et al., 2013. En étudiant avec attention la littérature de ces dernières années, on constate que de nombreux travaux traitant de la simulation numérique du couplage entre les phénomènes de transfert par diffusion des ions chlorures et les mécanismes de fissuration sont apparus. Kamali-Bernard et Bernard [Kamali-Bernard et Bernard, 2009] ont étudié l'influence de fissures obtenues par un essai de traction sur la diffusivité de l'eau dans le mortier. Dans leurs travaux numériques basées sur la méthode EF, Jin et al. [Jin et al., 2008], ont pris en compte la fissuration dans le coefficient de diffusion en se basant sur les travaux de Djerbi et al. [Djerbi et al., 2008]. Leurs résultats numériques ont été comparés avec les résultats expérimentaux obtenus par Ismail et al. [Ismail et al., 2008]. Dans le travail de Jin et al. [Jin et al., 2008], seule une fissure artificielle est considérée. Aucune simulation mécanique n'est effectuée pour obtenir la fissure. L'orientation et la position de la fissure dans le maillage sont imposées par l'utilisateur, en tenant compte d'une largeur, d'une hauteur et d'un angle donnés. Wang et Ueda [Wang et Ueda, 2011] ont considéré un modèle de type treillis à l'échelle mésoscopique tri-phasique (ITZ, agrégats, mortier) pour évaluer l'influence de l'interface ITZ et des agrégats sur la profondeur de la pénétration de chlorures. Encore une fois, dans [Wang et Ueda, 2011], seule une fissure artificielle est prise en compte. Savija et al. [Savija et al., 2013, Savija et al., 2014] ont simulé la diffusion d'ions chlorure dans le béton fissuré avec un modèle mésoscopique en treillis considérant le béton comme un domaine hétérogène incluant les agrégats, la pâte de ciment et l'auréole de transition (ITZ). Contrairement aux travaux numériques précédents, des simulations mécaniques sont réalisées, menant à des fissures réalistes (non artificielles).

Dans ce travail, la Embedded Finite Element Method (E-FEM, Armero et Garikipati, 1996], [Oliver, 1996], [Borja, 2000], [Benkemoun et al., 2010]) est introduite pour permettre de prendre en compte l'hétérogénéité du béton à l'échelle mésoscopique. L'avantage de cette méthode est de conserver tous les points forts de l'approche des éléments finis (applicabilité pour différentes lois constitutives, robustesse, compatibilité avec les techniques d'homogénéisation, ...) tout en facilitant considérablement l'étape de maillage des discontinuités physiques telles que les fissures, les fractures et les interfaces du matériau. En effet, le maillage n'a pas nécessairement besoin de suivre ces discontinuités physiques. Elles sont prises en compte en enrichissant l'espace d'approximation des éléments finis grâce à des améliorations spécifiques dans les champs de déplacement/contrainte pour les problèmes mécaniques et/ou dans le gradient de pression [Alfaiate et al., 2010] et le gradient de température [Ngo et al., 2013] pour les problèmes multi-physiques, par exemple. Dans ce cas, avec l'Embedded Finite Element method, le maillage n'a pas besoin de suivre la discontinuité physique qui est définie ici comme l'interface du matériau séparant la matrice du mortier et l'agrégat. Par conséquent, certains éléments finis sont traversés par cette interface et sont donc divisés en deux parties ayant chacune différentes propriétés de diffusivité. Afin de tenir compte de cette interface dans ces éléments, l'espace des éléments finis est enrichi par une discontinuité faible, située à l'interface des deux matériaux. Cette discontinuité faible est introduite dans la cinématique des champs réel et virtuel de concentration de chlorures libres.

Concernant les travaux de simulations dans la littérature, les modèles numériques capables de fournir des informations macroscopiques de transport tenant compte de la fissuration et de l'hétérogénéité du matériau du béton restent peu abordés. Par conséquent, nous proposons dans ce travail une méthode reposant sur un processus d'homogénéisation dont le but est de fournir une diffusivité macroscopique (tenseur de diffusion moyen) prenant en compte le caractère hétérogène du béton, le mécanisme de diffusion, les interactions chimiques (entre les chlorures et la matrice cimentaire) et intégrant le niveau d'ouverture de fissure et le chemin de fissuration. Dans ce cas, les points clés du processus de fissuration comme l'évolution d'une fissuration répartie vers une fissuration localisée (macro-fissure(s)), la tortuosité de la fissure et l'anisotropie induite par la fissuration sont ainsi intégrés naturellement dans la diffusivité macroscopique.

Objectifs de la thèse

Les principaux objectifs de la présente étude sont résumés comme suit :

- Décrire brièvement le rôle crucial de la pénétration des ions chlorure et son influence sur la durabilité des structures en béton armé et en particulier la corrosion des armatures.
- 2. Étudier l'influence des propriétés de la microstructure ainsi que la capacité de fixation chimique des chlorures par la matrice cimentaire sur les profils de concentration en chlorure dans le cadre d'une simulation en 1D.
- Effectuer des études d'homogénéisation numériques sur les méso-structures en 3D afin de calculer les tenseurs de diffusivité macroscopiques tenant compte du matériau bi-phasique non fissuré.
- 4. Introduire un couplage entre le transfert ionique par diffusion et la fissuration mécanique à l'échelle mésoscopique pour la détermination de tenseurs de diffusivité macroscopique dans des matériaux quasi fragiles, hétérogènes tels que le béton.
- 5. Étudier l'effet la fixation de chlorures ainsi que la diffusivité induite par la fissuration sur la durabilité et la prédiction de la durée de vie.

Organisation de la thèse

Ce mémoire est composé de quatre chapitres : Le premier présent une revue de la littérature. En général, il contient une revue des mécanismes du transfert ionique et de la corrosion des armatures due aux attaques des chlorures venant des eaux de mer et des sels de déverglaçage. Ensuite, les modèles numériques de couplage entre le transfert ionique par diffusion et endommagement mécanique utilisés dans la littérature sont décrits et commentés.

Le deuxième chapitre décrit les concepts de base de la méthode des élément finis utilisée pour résoudre un problème unidimensionnelle de transport. Aussi, nous présentons la Embedded Finite Element Method (E-FEM) tenant compte de l'hétérogénéité du béton à l'échelle méso. Ceci est obtenu en introduisant une discontinuité faible (dans le champ de concentration de chlorures) afin de capturer les hétérogénéités. La validité de la formulation EF implantée dans le code d'éléments finis FEAP [Taylor, 2008] est confirmée par des solutions analytiques unidimensionnelles pour un matériau semi-infini et bi-phasique. Tout d'abord, il est utilisé pour examiner l'impact des valeurs du coefficient de diffusion des agrégats et de la matrice de mortier ainsi que la fixation d'ions chlorure sur le profil de concentration. Une fois que l'évolution des profils de concentration pour différents intervalles de temps est connue, l'intervalle de temps jusqu'à l'initiation de la corrosion t_{corr} peut être évalué. Ainsi, t_{corr} se trouve :

- diminué lorsque les effets de la capacité de fixation au chlorure sont pris en compte;
- augmenté lorsque le rapport entre les coefficients de diffusion de la matrice et des agrégats $D_{matrice}/D_{aggregate}$ accroit.

De toute évidence, afin de mieux prévoir le temps d'initiation à la corrosion, il est important de tenir compte explicitement de l'influence des propriétés de la microstructure et de la capacité de d'interaction chimique de la matrice cimen-

taire sur la pénétration de chlorures. Ceci est crucial pour une estimation précise de la durée de vie d'une structure. Dans le chapitre 3, en effectuant des simulations en 3D, nous montrons que la méthode d'homogénéisation retenue dans ce chapitre combinée à l'E-FEM est bien adaptée au calcul des tenseurs de diffusivité macroscopiques prenant en considération des propriétés de diffusivité de la microstructure. Des simulations numériques sur des échantillons 3D sont effectuées pour déterminer la taille du VER. La précision des E-FEM pour le calcul du tenseur de la diffusivité à l'échelle mésoscopique est prouvée en comparant les résultats numériques aux résultats de l'équation de Maxwell. La comparaison avec des résultats expérimentaux est également effectuée pour montrer l'applicabilité de l'approche numérique proposée concernant l'effet des interactions chimiques. Dans le chapitre 4, le modèle mécanique utilisé pour prendre en compte la fissuration, est brièvement présenté dans un premier temps. En plus de la discontinuité faible, une discontinuité forte est également introduite pour tenir compte de la microfissuration. Dans un deuxième temps, le couplage entre le transfert par diffusion et la fissuration mécanique est présenté. Ceci repose sur les travaux expérimentaux de Djerbi et al. [Djerbi et al., 2008]. Ensuite la méthode d'homogénéisation permettant le calcul du tenseur de diffusivité macroscopique est détaillée. Par ailleurs, un exemple numérique méso-macro est illustré. L'anisotropie induite par la fissuration et la tortuosité du chemin de la macro-fissure sont les principales caractéristiques considérées ici. Nous avons montré que le tenseur de diffusivité macroscopique intègre l'évolution de la fissuration. Une augmentation des valeurs des composantes diagonales du tenseur de diffusivité macroscopique est ainsi mise en évidence. Enfin, nous avons réalisé la prédiction de la durée de vie des structures en béton armé. La sensibilité des valeurs du tenseur de diffusivité macroscopiques sur l'estimation de la durée de vie des structures en béton, y compris l'effet de la fissuration et de la structure interne à l'échelle mésoscopique sont explicitées.

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List of Abbreviations

Symbol	Meaning
J	Diffusive mass flux of chloride ions
D_e	Effective diffusion coefficient
С	Mass concentration
C_S	Concentration at the exposure surface
<i>C</i> ₀	Initial concentration in the specimen
erf()	Standard error function
Z	Ion valence
F	Faraday constant
R	Gas constant
Т	Absolute temperature
$ \triangle E $	Potential difference applied across the sample
L	Specimen length
k	Intrinsic permeability of the porous medium
ν	Dynamic viscosity of the fluid
$\mathbf{\nabla}p$	Pressure gradient
D_h	Humidity diffusion coefficient
C_t	Total chloride concentration
c_f	Free chloride concentration
c_b	Bound chloride concentration
α_L	Binding Langmuir isotherm parameter
β_L	Binding Langmuir isotherm parameter
D_c^*	Apparent diffusion coefficient

Symbol	Meaning
D_{cr}	Diffusion coefficient through the crack
[u]	Crack width
D_o	Diffusion coefficients in free solution
B_{cr}	Crack geometry factor
D_a	Chloride diffusivity of aggregate
D_p	Chloride diffusivity of cement paste
$\vec{q}(\vec{x})$	Mass flux
D_m	Mesoscale diffusion coefficient
$\vec{\nabla}$	Gradient operator
$c(\vec{x})$	Mass concentration
\vec{G}	Mean concentration gradient
$ec{Q}$	Mean flux gradient
V	Volume of Ω
L_s	Sample size
$ \bullet _F$	Frobenius norm
$\underline{\underline{A}}_{\underline{\underline{D}}}$	The anti-symmetric part of $\underline{\underline{D}}$
$\underline{\underline{S}}_{\underline{\underline{D}}}$	The symmetric part of $\underline{\underline{D}}$
$G_1^{(1/2)}$	Scalar function (first incompatible mode)
heta	Non-dimensional function to position the interface
δ_{Γ}	Dirac generalized function placed at the interface
G_2	Second incompatible mode allowing to incorporate the cracking
Φ	The yield function

Symbol	Meaning
t_{Γ}	Traction vector over the discontinuity
q	Stress like variable
G_f	Fracture energy
$ ilde{\epsilon}_1 \;, ilde{\epsilon}_2$	The enhanced strain fields
$\nabla^s u$	Displacement gradient
t_{\circ}	Corrosion initiation time
t_p	Corrosion propagation time
t_{cr}	Critical time at which corrosion starts to induce cracks
C_{th}	Threshold chloride concentration
d_c	Concrete cover
$ ho_{cor}$	Corrosion product density
d_s	Diameter of the bar
S	Space between steel bars
j_r	Rust production rate
δ_{pp}	Bar hole flexibility

General introduction

0.1 General

Reliable methods for ensuring and evaluating sustainable development of constructions and infrastructure are essential in the civil engineering industry, specifically in terms of durability. In order to improve durability, two approaches are needed: (1) a better understanding of the long-term behavior of cement-based materials subjected to various aggressive environmental conditions, and (2) an assessment of the mechanisms involved in their degradation.

In reinforced concrete civil engineering facilities, a number of external corrosive agents, such as dissolved salt in sea water and spray, and deicing salt spread on roads in winter, as well as in the constituents of concrete (sand or additives) can penetrate through cement materials. Chloride ions are the most corrosive agents with regard to concrete. Once they have penetrated the concrete cover, chloride ions may infiltrate the reinforcement, which can become corroded when the threshold value is reached [Mehta and Monteiro, 2006].

Consequently, mechanical performances are deteriorated resulting in a severe impact on durability and service life of the structure. The penetration rate of chloride ions is extremely dependent on the diffusion properties of concrete constituents. In addition, cracking can considerably accelerate the ingress of chloride ions inside concrete due to the fact that it provides preferential flow channels and enable additional chlorides to penetrate. Therefore, modeling cracking while explicitly tacking into account the different constituents of concrete is a challenging issue for a realistic prediction of the diffusivity of concrete.

Concerning the different constituents of concrete and in a more general way, the level of heterogeneity retained for the modelling of concrete-like material, several different representative scales can be regarded for analyzing the impact of this level of heterogeneity: macro, meso, micro and recently also the nano-scale are worth noting.

On the one hand, in macroscopic models, concrete is generally considered as homogeneous material and its behaviour is characterized using phenomenological laws such as plasticity and damage [Lemaitre and Chaboche, 2004]. These models are used for instance for structural analysis and design of a global concrete structure. Nevertheless, the macroscopic scale does not explicitly take into account the impact of the microstructure and properties of the individual constituents of concrete. Consequently, physical mechanisms (cracking, degradation or transport mechanisms) taking place at the fine scales are not captured in these macro-models and responsible of what it is observed at the upper scale (such as the macro one). On the other hand, nano and microscopic scales could give an accurate description of the different constituents and their physical behaviour. Nevertheless, the complexity of the different phases and also their large number make the calculations more time-consuming and the identification of the material parameters quite complicated.

At the mesoscopic scale, the heterogeneities are less marked: concrete is com-

monly regarded as coarse aggregates embedded in a mortar matrix. In this sense, mesoscopic models can be used to characterize quite precisely the material by providing an explicit representation of the heterogeneity (shape, distribution, size and behaviour) present at the fine scales while keeping a resaonable time of computation. In addition, since the pioneering works of Schlangen and van Mier Schlangen and van Mier, 1992, modeling a material using mesoscale models that explicitly take into account the microstructure of the specimen has become increasingly popular. These models have proved to be effective in modeling both the mechanical features [Benkemoun et al., 2010, Roubin et al., 2015a, Comby-Peyrot et al., 2009] and the mass transport aspect [Jourdain et al., 2014b, Benkemoun et al., 2015, Grass] and Bolander, 2016) of concrete-like materials. Consequently, we turn to the use of meso-scale model in this PhD. Concerning diffusion problem solved in the context of meso-scale approach, we can note the following numerical contributions. Garboczi and Bentz [Garboczi and Bentz, 1998] suggested a comprehensive multiscale model to predict the diffusivity of concrete by taking into account the effect of the Interfacial Transition Zone (ITZ). Zheng et al. [Zheng et al., 2012] worked with the lattice model to evaluate the chloride diffusion in concrete according to the simulated meso-structure of concrete. In addition the effect of aggregate shape on chloride diffusivity was examined. Zeng [Zeng, 2007] utilized the finite element method with a two-dimensional structure model, to analyze the chloride diffusivity behavior in concrete. The author simulated concrete as a two-phase composite made of aggregate and mortar matrix. Li et al. [Li et al., 2012] used also the finite element method to predict the behavior of chloride diffusivity in concrete with two and three-dimensional model and as a two-phase composite. The effect of ITZ was considered to estimate the chloride diffusion coefficient at mesoscale by Abyaneh et al. [Abyaneh et al., 2013]. Wang and Ueda [Wang and Ueda, 2009] employed a two-dimensionnal mesoscale truss network model for estimating chloride diffusion in concrete and considering the effect of time and temperature [Du et al., 2014].

However, chloride ions transport in concrete is generally not a simple diffusion process but a mass transport associated to different phenomena such as migration and also chloride dissipation throughout diffusion process due to chloride binding. Many researches conducted the study of chloride ions diffusivity in cement-based material under the effect of different transport mechanisms (permeability, migration, chloride binding). For instance, a meso-scale truss network model was used by Wang and Ueda [Wang and Ueda, 2009] to investigate chloride diffusion considering concrete as a three-phase composite of mortar matrix, aggregates, and the interfacial transition zone (ITZ). Diffusion law is considered in this work taking into account the dependency of diffusion coefficient of mortar and ITZ on exposure duration and temperature. The same model was utilized by Wang and Ueda Wang and Ueda, 2011b] to evaluate the diffusivity of concrete in order to account for the microstructure of concrete, the binding effect of chloride ions and the chloride concentration dependence. Note that in both aformentioned works of Wang and Ueda, the chloride diffusivity of mortar and ITZ are estimated by analytical method. The numerical results show the importance of considering the binding capacity and concentration dependence on chloride ions transport modelling. A Finite Element method for numerical solution of the Poisson-Nernst-Planck equations was introduced by [Krabbenhøft and Krabbenhøft, 2008] to model the migration test. Then, closed-form solutions for the effective chloride diffusivity based on the full

Poisson-Nernst-Planck equations are derived. However, chloride binding was taken into account in the work of Zeng [Zeng, 2007] where a Finite Element method was used to investigate the chloride diffusion behavior of a two-phase concrete material. The results show that chloride diffusion behavior at the level of heterogeneity is different from that in its derived homogeneous medium.

As aforementioned, modeling chloride ions transport is only reliable if it takes into account the principal issue in the degradation of concrete structures, i.e., cracking. Cracking is responsible [Djerbi et al., 2008] for structure-accelerated damage when localized cracks (such as macrocracks) appear [Djerbi-Tegguer et al., 2013].

In the last few years, some research works have been reported in the literature on the numerical simulation of the coupling between chloride ions transport and concrete cracking. Realistic numerical studies are essential because it is extremely difficult to set up an experimental investigation of this coupling. Herein, considerable computational work has been done. Kamali-Bernard and Bernard [Kamali-Bernard and Bernard, 2009] investigated the influence of tensile cracking on diffusivity of tritiated water in mortar within a numerical microstructure model. In Jin et al. [Jin et al., 2008], the authors integrate the diffusion coefficient through the crack obtained in Djerbi et al. [Djerbi et al., 2008] in their numerical model of diffusion based on the Finite Element method. Jin et al. [Jin et al., 2008] compare their numerical results with the experimental ones of Ismail et al. [Ismail et al., 2008]. Note that in the work of Jin et al. [Jin et al., 2008], only artificial crack is considered. The term "artificial" means that no mechanical simulations are performed to obtain the crack. The orientation and position of the crack in the mesh are imposed by the user, considering a given width, height and angle. Wang and Ueda [Wang and Ueda, 2011a] consider a mesoscale lattice-type model with three phases (ITZ, aggregates, mortar) to assess the influence of the ITZ and the aggregates on the depth of chloride penetration. Here again in [Wang and Ueda, 2011a], only artificial crack is taken into account. Šavija et al. [Savija et al., 2013, Savija et al., 2014] simulated the chloride ions diffusion in cracked concrete with a coupled lattice mesoscale model considering concrete as heterogeneous domain including aggregates, cement paste and interfacial transition zone (ITZ). Contrary to the previous mentioned works, mechanical simulations are performed leading to realisitic cracks. In Šavija et al. [Savija et al., 2014], according to findings of Yoon et al. [Yoon et al., 2007], the authors impose a chloride concentration only on cracks wider than 12 μ m obtained after the numerical simulation of a splitting test. In Šavija et al. [Savija et al., 2013], the authors assess their numerical model with (1) the experimental results of Sahmaran [Sahmaran, 2007] and Ismail et al. [Ismail et al., 2008] for cracked mortar samples by considering D_{cr} of Djerbi et al. [Djerbi et al., 2008], (2) the experimental results of Ismail et al. [Ismail et al., 2004a] for cracked bricks samples – using trial and error method to obtain the equivalent diffusion coefficient – in the context of artificial cracks. Wang et al. [Wang et al., 2008] presented mesoscale model to simulate chloride diffusivity in cracked concrete using the Rigid Body Spring Model and the truss network model. Rigid Body Spring Model (RBSM) is employed to perform the mechanical analysis to simulate the distribution and width of microcracks. Then, the truss network model is utilized to assess the chloride diffusivity of the cracked The axial compressive and tensile loading conditions are investigated concrete. respectively and the effects of stress level on chloride diffusivity of cracked concrete are studied.

Regarding the FE literature, the interest of the computational mechanics community has significantly increased over the last decades in the use of Enriched Finite Element Methods such as the Generalized Finite Element Method (G-FEM, [Strouboulis et al., 2000a], [Strouboulis et al., 2000b]), the eXtended Finite Element Method (X-FEM, [Belytschko et al., 2001], [Moës et al., 1999], [Mariani and Perego, 2003), the Nitsche method ([Hansbo and Hansbo, 2002] and [Annavarapu et al., 2012) and the Embedded Finite Element Method (E-FEM, [Armero and Garikipati, 1996], [Oliver, 1996], [Borja, 2000], [Benkemoun et al., 2010]). The advantage of these methods is to retain all the advantages of the finite element approach (applicability to different constitutive laws, robustness, compatibility with homogenization techniques, ...) while considerably easing the meshing step of physical discontinuities such as cracks, fractures, slip lines and material interfaces. Indeed, the mesh does not necessarily need to match these physical discontinuities. They are taken into account by enriching the finite element approximation space through specific enhancements in the displacement/strain fields for mechanical problems and/or in the pressure gradient [Alfaiate et al., 2010] and temperature gradient [Ngo et al., 2013] for multi-physics problems for instance. Consequently, these physical discontinuities have the representation of their (complex) geometry and its evolution through the time facilitated by means of these methods. In the literature, the enhancements are in general sorted into two categories:

- strong enhancements a jump within the displacement (and/or the pressure, the temperature, the concentration,...) fields is introduced by the means of a so-called strong discontinuity ([Simo et al., 1993]);
- weak enhancements a jump within the strain (and/or the pressure gradient,

the temperature gradient, the concentration gradient,...) fields is introduced by the means of a so-called weak discontinuity ([Ortiz et al., 1987]).

Consequently modeling chloride ions transport with a meso-scale model based on the aforementioned Enriched Finite Element Methods might provide yet another efficient and original way to tackle durability issues relative to chloride ingress in sound and cracked concrete. It can be an efficient alternative numerical approach for taking into account the microstructure and cracking in the design of mesoscale models for chloride ions transport. In this PhD thesis, among the different Enriched Finite Element Methods evocated in the introduction, we turn to the Embedded Finite Element Method (E-FEM)

We have to note that macroscopic diffusivity tensors, which depend on the cracked state of concrete and its overall heterogeneous aspect, have rarely been computed in the numerical simulation literature. Some explanations for this could be: (1) by not considering realistic cracks, the authors avoid the connectivity and tortuosity aspects of the crack pattern and crack width heterogeneities in the computation and/or (2) the authors use an equivalent diffusion coefficient D_e , which can be considered a phenomenological variable such as damage or plasticity in mechanics [Reynouard et al., 2009]. However, D_e does not take into account the crack pattern failure or the heterogeneous aspect of the material explicitly. Clearly, these macroscopic tensors are critical in terms of the durability and service life of civil engineering facilities.

Having at hand, an homogenization technique and the Embedded finite element formulation for mesoscale chloride ions diffusion transport modeling, we estimate the macroscopic diffusivity tensors taking into account for the heterogeneous mesoscale structure of sound and cracked concrete, diffusion mechanism of transport and also chloride binding phenomena. This is the main originality of my thesis.

The following are the main aims of the present study:

- 1. To briefly describe the crucial role of chloride ions penetration on the corrosion of reinforcing steel in civil engineered facilities.
- 2. To investigate through 1D simulations the influence of the microstructure properties and the chloride binding capacity on the chloride concentration profiles.
- 3. To perform numerical homogenization experiments on 3D meso-structures to compute macroscopic diffusivity tensors accounting for two-phase material.
- 4. To introduce a cracking/mass transport mesoscale coupling for the determination of macroscopic diffusivity tensors in heterogeneous quasi-brittle materials such as concrete.
- 5. To investigate the influence of chloride binding contribution and also the cracking-induced diffusivity on the durability and service-life prediction.

To reach these objectives, this thesis is outlined in four chapters.

In Chapter 1 a comprehensive literature review is presented. It contains a review about chloride ion transport mechanisms and reinforcement corrosion due to chloride attack. A survey of the literature is then established to show the relevant transportcracking coupling models.

Chapter 2 describes the basic concepts of the FE method to carry out a one dimensional solution of a transport problem to better understanding the formulation and the implementation of the FE solution code. While, in section (2-4) of this chapter, we turn to the Embedded Finite Element Method (E-FEM) in order to take into account the heterogeneity of concrete at the mesoscale. The influence of the microstructure properties and the chloride binding capacity on the chloride concentration profiles and consequently on the corrosion initiation time is investigated. This chapter is limited to 1D description.

Chapter 3 contains the 3D implementation of the computational homogenization retained in this chapter combined to the E-FEM to compute macroscopic diffusivity tensors considering the microstructure diffusivity properties. Herein, chloride binding is taken into account.

In chapter 4, in a first step, the mechanical model used to carry out the mechanical calculations is briefly presented and Key points are also highlighted. In a second step, the coupling between diffusion transfer and mechanical cracking is presented. It is based on the experimental work of Djerbi et al. ([Djerbi et al., 2008]). Then, the homogenization method for calculating a macroscopic diffusivity tensor is detailed. A meso-macro numerical example is presented to carry out the computation of macroscopic crack-induced diffusivity tensors. Then, the performance of civil engineering facilities through the investigation of service life prediction is quantified.
Chapter 1

Literature review

1.1 Introduction

In the field of Civil Engineering, establishing a methodology for sustainable development, which integrates the durability of buildings or civil engineering infrastructures is essential today. This durability requires (1) a better understanding of long-term behavior of cement-based materials in various aggressive environments, and (2) an evaluation of the associated degradation mechanisms (aging, loss of bearing capacity and risk of collapse, ...). One of the most complex attacks on any reinforced concrete structure during its service life can be induced by chloride ingress.

There are two primary sources of chloride ions attacks referred as: external or internal. The external attack could come from seawater weathering or de-icing salt, while the internal one could arise when water used during concrete mixing contains chlorides or sometimes certain additions of chloride-based admixtures. External sources of chlorides are usually responsible for the initiation of corrosion while the effect of internal chlorides is often negligible [Jefremczuk, 2004].

When the chloride ions penetrate gradually into reinforcing steel cover, they can reach the reinforcements, destroy the passive layer that protects the reinforcements and corrode them [Mehta and Monteiro, 1993], consequently, the durability of the infrastructure can be affected by a loss of its mechanical performances.

Modelling of different transport mechanisms -diffusion, migration, and convection- of chloride ions and the behavior of these ions -interactions with other ions- in a cementitious material -saturated or unsaturated- was the subject of numerous studies. However, this modeling of chloride ions transport is more realistic if it integrates the main degradation mechanism of concrete structures in service, namely cracking. Indeed, as the pores of the material, cracking is responsible for the chloride ions penetration and can cause an accelerated corrosion of steel reinforcement, since they may represent a favorable path for the aggressive agents.

In the literature, some research work has been devoted to studying the transfer of chloride ions in sound (uncracked) concrete. However, to approximate real loading conditions where the ions' transfer is done in the structural elements (columns, beams), undergoing mechanical loads cracking, it is important to study the coupling between cracking and chloride ions transport. In this field, most of the previous studies are limited to Fick's law for the transfer modelling.

The main objectives of this chapter are: to briefly describe the crucial role of

chloride ions penetration on the corrosion of reinforcing steel in civil engineering facilities. The basic principles and existing models of chloride ingress are shown as well. A survey of the literature is established to light on the relevant transportcracking coupling models.

1.2 Corrosion of reinforcement

The normal concrete has a highly alkaline pore solution. At such alkalinity in the case of reinforced concrete structures, steel is normally protected against corrosion by a thin passive oxide film. Steel is said to be depassivated if the stability conditions of the protective layer are changed, the state of passivation ceases and reinforcement corrosion starts. The enormous expansion of rust products could increase the internal stresses of concrete elements to cause firstly cracking of the concrete skin. Two other harmful effects on the performance of the reinforced concrete structure can occur: a loss of steel section combined with a loss of steel-concrete bond.

Two external agents contribute to the depassivation of reinforcement: carbon dioxide and chloride ions. Hereafter, the next section presents the principal mechanism that may induce corrosion of reinforcing steel by chloride ions ingress and a description of the corrosion process. The carbonation is out of the domain of this study. Chloride induced reinforcement corrosion is formed in three steps: penetration of chloride ions, depassivation, corrosion initiation and propagation.



Figure 1.1: Schematic representation of pitting corrosion of steel in concrete, Hassan Z. F. [Abu Hassan, 2012]

1.2.1 Penetration of chloride ions

Chloride ions present in the concrete can come from two different sources. Either when water used during concrete mixing contains chlorides or sometimes certain additions of chloride-based admixtures. Or from the external environment such as marine or deicing salts NaCl and $CaCl_2$), which diffuse into the concrete.

After the entrance of chloride ions either during the mixing process or from the exterior sources such as sea water or deicing salts, chloride ions in concrete exist in two forms: free chlorides which are dissolved in the pore solution or chlorides bound to hydrated cement or adsorbed on the surface of the pore walls. Free chloride ions can migrate through concrete pores, under the effect of concentration gradient between the inside and the outside or under the effects of other governing mechanisms such as capillary suction or hydrostatic pressure.

The transport of these ions is related to the porosity of the concrete. It decreases with the W/C ratio and depends on the concentration of the pore solution salts.

1.2.2 Depassivation

The immunity of reinforcement is ensured by a kind of passive film Fig.(1.2) which is commonly formed in a highly alkaline environment with a high pH value of about 12.5-13.5 to protect the steel from active corrosion [Hansson et al., 2007]. The process of corrosion begins after the concrete cover being penetrated by



Figure 1.2: steel reinforcement protected by a passivating layer

the last mentioned ions, in the conditions of the chloride content increase at the concrete-steel interface and reaches a critical threshold chloride concentration causing the active corrosion to be triggered. The methods of determining this critical chloride threshold concentration are varied.

It is hard to specify in a convincing manner the critical concentration capable of initiating reinforcement's corrosion. The influence of different factors such as quality of concrete, composition of concrete, W/C ratio, relative humidity, temperature, the microstructure in contact with the reinforcement, the amount of present sulfate in concrete, and the steel surface condition all are factors that have an impact on this parameter [Rafiee, 2012].

Steel corrosion is observed in the presence of an enormous concentration of chloride ions in the concrete. They alter the morphology of the passive layer giving ions $FeCl_3^-$ or $FeCl_2$. The corrosion products produce a volume several times greater than the initial steel bar's size.

1.2.3 Corrosion initiation and propagation

The corrosion-induced deterioration of reinforced concrete can occur in two phases (see Fig.1.3) :



Figure 1.3: Service life of concrete structure subjected to corrosion [Tuutti, 1980]

Initiation phase: corresponds to the slow chemical and physical changes of the concrete cover at the micro-scale. It starts from the time where aggressive species reach reinforcement surfaces and trigger active corrosion, and ends once the corrosion products starting to accumulate in the concrete-steel interface.

Propagation or development phase: where the material degradation is visible. The steel bars will be covered by intermediate products and consequently concrete cover is damaged. These products have a volume of 2 to 8 times that of steel bar [Mehta and Monteiro, 1993]. A further rust accumulation will trigger expansive tensile stress about 10 times the tensile strength of normal concrete and arise up to 27.5 MPa and then cracking in the surrounding concrete [Chen and Mahadevan, 2008]. The result of damage of concrete cover is a significant deterioration in the load carrying capacity of concrete structure element.

The second dangerous effect of corrosion is the progressive reduction of the cross-section of steel bars and thereby decreasing the ability to withstand the applied service loads leading to failure of the structure [Al-Kutti, 2011]. A schematic sketch of the chloride induced corrosion process is shown in Figure 1.4. The period of this phase depends principally on the corrosion rate, which is affected by several factors [Otieno et al., 2008].



Figure 1.4: A schematic sketch of the chloride induced corrosion process [Rafiee, 2012]

1.2.4 Threshold Chloride Content

The chloride threshold level can be defined as the required concentration of chlorides in order to destroy the protective oxide film on surrounding the steel rebars and trigger the corrosion [Otieno et al., 2008]. Critical chloride concentration classified as one of the crucial input criteria for design requirements and service life evaluation to assess the time needed to reach the chloride threshold value at reinforcement level.

Several works have proposed various approaches to reveal the chloride threshold levels consisting of: free chloride threshold level, total chloride, and $[Cl^-]/[OH^-]$. According to Angst et al. [Angst et al., 2009], threshold values range from 0.04 wt% to 8.34 wt% of cement. Many reasons are responsible for this large variation of threshold value such as: type of steel, concrete quality, cater/cement ratio, etc.

1.3 Mechanisms of chloride transport into concrete

Today, there are increasingly premature degradation problems on reinforced concrete structures in contact with aggressive agents such as seawater or deicing salts. Several different mechanisms control the chloride ions transport into concrete such as diffusion, permeation, migration and convection (or capillary suction) according to the surrounding conditions as shown in Fig.(1.5) (see [Poulsen and Mejlbro, 2010] for details).

In saturated parts of concrete structures in marine environment, diffusion is the

main mechanism controlling the chloride ingress in marine environment, while the convection could be ignored [Al-Kutti, 2011]. In the case of non-saturated parts of concrete structures, chloride ions are mainly absorbed into concrete pore by capillary suction mechanism [Djerbi et al., 2008]. In practice, any (combination) of these mechanisms can govern the ingress of chloride ions into concrete according to the surrounding conditions (see Fig. 1.5).



Figure 1.5: Exposure zones of concrete structure for marine environment, Ghods et al. [Ghods et al., 2005]

1.3.1 Diffusion

Diffusion is the prevailing transport mechanism of chloride in concrete. It based on mathematical model formalized by Adolph Eugen Fick (1829-1910). It can be defined as the movement of free ions from the region of higher concentration to region of lower concentration. Such conditions are encountered for instance in the submerged zone and the tidal zone (high tide) of structures under marine environment. The debut of employing of Fick's second law of diffusion in the transport of chloride ions problem had appeared, in the first time, through the works of Mario Collepardi in 1970's [Collepardi et al., 1970, Sillanpää, 2010].

Hence, in steady state flow, mass flux obey Fick's first law of diffusion (see Fig. 1.6):

$$\boldsymbol{J} = -\boldsymbol{D}_e \,\,\boldsymbol{\nabla}\boldsymbol{c} \tag{1.1}$$

where J the diffusive mass flux of chloride ions expressed in $[kg/m^2.s]$, D_e the effective diffusion coefficient $[m^2/s]$, and c the mass concentration $[kg/m^3]$. Mass



Figure 1.6: Conservation of mass

balance for chloride ions can be expressed as:

$$\frac{\partial c}{\partial t} + div \boldsymbol{J} = 0 \tag{1.2}$$

Injecting equation 1.1 in the mass balance equation [Poulsen and Mejlbro, 2010],

Ficks second Law is obtained such as:

$$\frac{\partial c}{\partial t} = D_e \frac{\partial^2 c}{\partial x^2} \tag{1.3}$$

In the case of constant chloride concentration of the exposure surface, constant diffusion coefficient D_e and semi-infinite space, Eq. 1.3 has an analytical solution (see [Crank, 1979]), such as:

$$c(x,t) = c_0 + (c_s - c_0) \left(1 - erf\left(\frac{x}{2\sqrt{D_e t}}\right) \right)$$
(1.4)

where $c_s = c(x = 0, t > 0)$ the concentration at the exposure surface, $c_0 = c(x > 0, t = 0)$ the initial concentration in the specimen, and erf() the standard error function [Zhang, 2013].

1.3.2 Migration

As well as being relatively complex, chloride ions diffusion through concrete is a slow process, and it usually takes several months to several years to reach the reinforcement. However, to get rid of this long time issue, accelerated techniques are often used, in the experimental setup, to measure chloride ions diffusivity in cementitious materials.

These accelerated methods consist in applying an electric field across a concrete specimen to speed up the transport of ions in the material and reduce the duration of the test. Within these conditions, the movement of the ions from areas of high electric potential to areas of low electric potential is governed by an external electric field and the ions transport is then defined migration [Arsenault, 1999].

Generally, there are several models found in literature used for interpreting the nonsteady state migration test. Most of these models relies on Nernst-Planck equation describing the migration of chloride ions in cementitious materials. Nernst-Planck equation (1.5) describes the flow of ions through a porous material [Nilsson et al., 1996], such as:

$$D_e \frac{\partial^2 c}{\partial x^2} - V D_e \frac{\partial c}{\partial x} - \frac{\partial c}{\partial t} = 0$$
(1.5)

The second term is the pure migration term caused by an electrical potential gradient.

$$V = \frac{zF|\triangle E|}{RTL}$$

where:

z: ion valence

F: Faraday constant

```
R: gas constant
```

T: absolute temperature

 $|\Delta E|$: potential difference applied across the sample

L: specimen length

In semi-infinite space where the boundary condition are :

$$t = 0, x > 0, c = 0$$

$$t > 0, x = 0, c = c_s$$

$$x \to \infty, c = 0$$

In the case of constant chloride concentration of the exposure surface, constant

diffusion coefficient D_e and semi-infinite space, Eq. 1.5 has an analytical solution as:

$$c = \frac{c_s}{2} \left(e^{Vx} erfc\left(\frac{x + VD_e t}{2\sqrt{D_e t}}\right) + erfc\left(\frac{x - VD_e t}{2\sqrt{Dt}}\right) \right)$$
(1.6)

where $c_s = c(x = 0, t > 0)$ the concentration at the exposure surface, $c_0 = c(x > 0, t = 0)$ the initial concentration in the specimen, and erf() the standard error function.

1.3.3 Permeation

The permeability can be defined as the capacity of a porous medium to transfer fluids -liquid or gas- under pressure gradient [Fenaux, 2013]. The parameter, to which permeability is sensitive, is the volume, distribution, connectivity, and tortuosity of pores and interconnected microcracks [Zhang, 2013].

The intrinsic permeability of the porous medium k (m^2) can be obtained by relying on the Darcy law for liquids, such as:

$$\boldsymbol{J_{Darcy}} = -\frac{k}{\nu} \boldsymbol{\nabla} p \tag{1.7}$$

Where J_{Darcy} the Darcy flux (m/s), k the permeability coefficient (m^2) , ν is the dynamic viscosity of the fluid (Pa.s) and ∇p the pressure gradient (Pa/m). Permeability is taken in consideration in water retaining structures like dams and other large hydraulic structures, where concrete structures being in contact with liquid under a relative high hydraulic pressure. [Otieno et al., 2008].

1.3.4 Convection

In the case of chloride ions transport in an unsaturated medium, two mechanisms govern the process: diffusion and absorption due to capillary phenomena between liquid and gaseous phases.

When concrete is subjected to repetitive wetting-drying cycles, the chlorides can penetrate into the concrete by capillary absorption and migrate through the liquid phase by advection within the material. In this case, ion transport is coupled with moisture transport. Then, convection can be defined as the transfer of dissolved chloride ions into pore solution by moisture flux [Guzmán et al., 2011].

This process occurs at a small distance from exposure surface causing inward and outward movement of moisture due to the wetting and drying cycles, respectively. As shown in Fig.1.7, this region can be defined as the convection zone. It is followed by the diffusion zone where diffusion being the governing mechanism of chloride ions movement [Bester, 2014]. Several authors proposed transport models



Figure 1.7: convection and diffusion zones [Bester, 2014]

to describe chloride penetration into partially saturated concrete. The global moisture transport in concrete can be expressed such as:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x} \left(D_h(\theta) \; \frac{\partial \theta}{\partial x} \right) \tag{1.8}$$

where θ the relative water content and D_h the humidity diffusion coefficient (m^2/s) . In the other hand, the governing equation of chloride ions transport in case of the unsaturated medium is defined such as:

$$\frac{\partial c_t}{\partial t} = \nabla \cdot \left(D_{cl}(\theta) \nabla c_f \right) + c_f \frac{\partial \theta}{\partial t}$$
(1.9)

where c_t the total chloride concentration and equal to the sum of free chloride concentration c_f and bound chloride concentration c_b and D_{cl} the chloride coefficient of diffusion as a function of relative water content [Ababneh et al., 2003, Wang et al., 2016b].

Injecting Equ.1.8 in Equ.1.9 leads to the general diffusion-convection equation:

$$\frac{\partial c_t}{\partial t} = \frac{\partial}{\partial x} \left(D_{cl}(\theta) \ \frac{\partial c_f}{\partial x} + c_f \frac{\partial}{\partial x} \left(D_h(\theta) \ \frac{\partial \theta}{\partial x} \right) \right)$$
(1.10)

1.3.5 Chloride Binding

Chloride ions existing in concrete can be mainly classified into free (or uncombined) chloride ions -dissolved in the pore solution of cement paste- and bound chloride ions. Bound chloride can be classified, in turn, to chemically-bonded chloride ions into cement hydration products to form Friedel's salt and physically-bound chloride

ions when it is absorbed by the cement gel surface [Yuan et al., 2009, Van Tran, 2008, Rafiee, 2012].

In consequence, the total bound chloride ions C_b is the sum of those chemicallybound and those physically-bound chloride ions. The total amount of chloride ions in concrete is therefore the sum of the bound and free chloride ions, such as:

$$c_t = c_b + c_f \tag{1.11}$$

Where c_t is the total chloride ions concentration $(kg/m^3 \text{ of concrete})$, c_b and c_f are the bound and free chloride concentration $(kg/m^3 \text{ of concrete})$, respectively.

For the initiation and propagation stage of steel corrosion of reinforced concrete structures, several reviews supposed that only free chloride can be related to corrosion process as due to its ability to penetrate deeper inside the concrete cover to reach the surface of embedded steel bars [Tuutti, 1982, Yuan et al., 2009]. However, chloride binding can act as a retarder agent of corrosion initiation since it decreases free chloride concentration at the steel bars level. Moreover, the influence of formation of Friedel's salt reduces the porosity of concrete cover and consequently, slows down the transport of chloride ions and decreases the probability of corrosion [Yuan et al., 2009].

This inhibiting role of chloride binding phenomenon show how crucial to consider it in chloride ions transport modelling, specifically with regards to acquiring a more realistic prediction of service life of concrete structures under chloride attack environment [Abu Hassan, 2012, Glasser et al., 2008].

The value of bound chloride ions c_b depends on the concentration of free chloride ions c_f . This relationship has been described in some of the literature by means of the so-called binding isotherm. In principle, we can identify three main models. These are: linear isotherm, Freundlich isotherm, and Langmuir isotherm [Luping and Nilsson, 1993, Baroghel-Bouny et al., 2012]

1.3.5.1 Linear Isotherm

Although many researchers have stated that the relationship between bound and free chloride is non-linear [Sergi et al., 1992a, Luping and Nilsson, 1993, Martin-Pérez et al., 2000], the linear isotherm is frequently used in material models. A linear binding isotherm was proposed by Tuutti [Tuutti, 1982] for free chloride ions concentration lower than 20 g/l, such as:

$$c_b = \alpha c_f \tag{1.12}$$

Where c_b is the bound chloride concentration, α is a constant and the binding capacity representing the slope of the dotted line in Fig. 1.8 $\frac{\partial c_b}{\partial c_f}$ is accordingly constant. However, some authors reported that linear isotherm might overestimate or underestimate the quantity of bound chloride when free chloride elevated or dropped highly [Delagrave et al., 1997, Nilsson et al., 1996].



Figure 1.8: Binding isotherms for concrete [Martin-Pérez et al., 2000]

1.3.5.2 Langmuir Isotherm

Langmuir chloride binding isotherm can be expressed as:

$$c_b = \frac{\alpha_L c_f}{1 + \beta_L c_f} \tag{1.13}$$

where α_L and β_L are constants related to the concrete binder composition and are obtained from linear [Sergi et al., 1992a] or non-linear [Yuan et al., 2009] regression analysis of the experimental data. Tang [Tang, 1996] showed that Langmuir isotherm can represent more precisely the monolayer adsorption -i.e the slope of isotherm curve has a tendency to approach zero at high concentration of free chloride ions at low amounts of free chloride -lower than 0.05 M- [Yuan et al., 2009].

1.3.5.3 Freundlich Isotherm

When free chloride concentration is in the range (0.01-1M), chloride binding can be described better by Freundlich isotherm as stated by Tang and Nilsson [Luping and Nilsson, 1993]. Freundlich isotherm can be expressed as:

$$c_b = \alpha_F c_f^{\beta_F} \tag{1.14}$$

Where α_F and β_F are constant parameters determined experimentally.

Different values of isotherm constants α and β for Langmuir and Freundlich binding isotherm have been resulted by different experimental work and different boundary conditions. Also, various units used for chloride ions concentration leads to various values of α and β , (see [Yuan et al., 2009] and [Martın-Pérez et al., 2000] for details). Many researchers have regarded that the apparent diffusion coefficient can represent the transport properties of chloride at pore solution taking into account the effect of chloride binding [Luping and Nilsson, 1993]. So the transport model based on Fick's 2nd law in one dimension can be adapted as:

$$\frac{\partial c_f}{\partial t} = \frac{\partial}{\partial x} \left(D_c^* \frac{\partial c}{\partial x} \right)$$

$$D_c^* = \frac{D_c}{1 + \frac{1}{\omega_e} \frac{\partial c_b}{\partial c_f}}$$
(1.15)

Where D_c^* is the apparent diffusion coefficient (m^2) and $\frac{\partial c_b}{\partial c_f}$ is the binding capacity which in turn presented as the slope of the related binding isotherm [Nilsson et al., 1996].

In previous sections, the equations of chloride transport in several conditions (saturated, accelerated,), binding isotherms are described. All of these mechanisms will be used in chapter 2 and 3 to show the methodology of modelling in (1D and 3D) and how the diffusivity tensor is effected in both sound and cracked concrete.

1.4 Transport/cracking coupling works

A considerable amount of effort has been devoted to the modelling of chloride ions transport mechanisms diffusion, migration, convection and behavior interaction or not with other ions, [Friedmann et al., 2004, Friedmann et al., 2008] within cement materials both saturated and unsaturated [Nguyen and Amiri, 2014, Nguyen and Amiri, 2016]. However, chloride transport modelling is reliable only if the main degradation factor of concrete structures in service, i.e., cracking, is considered. Cracking becomes the responsible of the accelerated penetration of chloride ions inside concrete and consequently the corrosion of reinforcement steel and material deterioration [Djerbi et al., 2008].

A survey of the literature reveals that during the last couple of years, some research work has been devoted to the numerical simulation of the coupling between chloride ions transport and concrete crack formation mechanisms. Numerous experimental studies were conducted to examine the effect of cracking on chloride ions transport or to assess the relation between the crack characters -such as crack width, depth and density- and material properties. Several testing methods have been followed for testing chloride transport in uncracked concrete basing on the different chloride transport mechanisms: diffusion, migration, capillary suction, etc. A number of such methods have been worked with to examine the effect of cracking on chloride ion ingress. The diffusion tests like NordTest NT Build 443 or steady-state diffusion test were used by several authors [Gu et al., 2015, Adiyastuti, 2005, Ismail et al., 2004b, Gowripalan et al., 2000, Rodriguez and Hooton, 2003]. However, short time migration tests such as NordTest NT Build 492 or its modified

method were used by other authors as a rapid method to figure out the chloride ion transport in concrete [Djerbi et al., 2008, Marsavina et al., 2009, Yoon et al., 2007].

In the other hand, Cracking can be formed in concrete as artificially or mechanicallyproduced. The term artificial means that no mechanical simulations are performed to obtain the crack. Artificial cracks were used by [Rodriguez and Hooton, 2003, Marsavina et al., 2009, Jin et al., 2010] where some factors influencing chloride penetration such as crack roughness, crack tortuosity and crack shape are not regarded for the sake of simplicity. The orientation and position of the crack in the mesh are then imposed by the user, considering a given width, height and angle.

Real cracks were induced by different methods and mechanical loading conditions such as tensile splitting test [Djerbi et al., 2008], four-point bending setup [Sahmaran, 2007], uniaxial compressive load [Wang et al., 2016a], etc. More details about testing the chloride penetration in cracked concrete can be found in [Gu et al., 2015, Blagojevic et al., 2012].

Concerning the coupling, researchers link an equivalent diffusion coefficient D_e to a crack width parameter which considered the main crucial factor found to change the material diffusivity of cracked concrete [Gu et al., 2015]. They propose models depending on the diffusion coefficient fitted from experimental data. In Jang et al. [Jang et al., 2011] and Djerbi et al. [Djerbi et al., 2008], a parallel model (accounting for a crack geometry factor in [Jang et al., 2011]) based upon the work of Gérard et al. [Gérard and Marchand, 2000] is set out. The model is then fitted on experimental results achieved from a migration test in steady-state regime on samples cracked by a tensile splitting test. Djerbi et al. [Djerbi et al., 2008] and Jang et al. [Jang et al., 2011] both suggest a linear relationship between the crack width and the equivalent diffusion coefficient D_e . In [Djerbi et al., 2008], an important step forward is made: the authors propose a relationship between the crack width and the diffusion coefficient through the crack D_{cr} . This information is of great interest for the mesoscale coupled approach detailed in this PhD thesis (see chapter 4). Within the predicted upper and lower threshold crack widths in [Djerbi et al., 2008], the relation between the diffusion coefficient through the crack widths in [Djerbi et al., 2008], the relation between the diffusion coefficient through the crack midth in [lu]] such as (see Fig. 1.9):

$$\begin{cases} D_{cr}(m^2/s) = 2 \times 10^{-11}[|u|] - 4 \times 10^{-10} \quad 30 \,\mu m \le [|u|] \le 80 \,\mu m \\ D_{cr}(m^2/s) = 14 \times 10^{-10} \quad [|u|] > 80 \,\mu m \end{cases}$$
(1.16)

The same approach is applied by Jang et al. [Jang et al., 2011]. The authors present



Figure 1.9: Effect of crack width on diffusion coefficient through the crack [Djerbi et al., 2008].

a quantitative relation between crack opening and diffusion coefficient considering a scalar factor accounting for tortuousity, connectivity and constrictivity of the crack which is called crack geometry factor. Therefore, the equivalent diffusion coefficient is:

$$D_{eq} = D + \frac{4\overline{w_{cr}}B_{cr}}{\pi d}D_o \tag{1.17}$$

where B_{cr} is the crack geometry factor ranged from 0.067 to 0.206. D_o , D_{cr} and D the diffusion coefficients in free solution, cracked and uncracked sections respectively, while d the specimen diameter. $\overline{w_{cr}}$ was introduced as the equivalent crack width to account for crack widths below the lower critical crack width.

In [Sahmaran, 2007], the author proposed an equivalent diffusion coefficient of a power function of the crack width. Here the fitting process relies on chloride concentration profiles. They have been obtained after that cracked samples with different level of crack width, achieved by a four-point bending setup, have been exposed to sodium chloride solution such as (see Fig.(1.10)):

$$D_{cr} = 3.46 + 0.0001975[|u|]^2, \quad [|u|] > 135\mu m \tag{1.18}$$

The authors consider the chloride flux as the total of flux in the cracked and uncracked partitions to evaluate the diffusion coefficient in cracked concrete as shown in Fig.(1.11)

In addition, Kwon et al. [Kwon et al., 2009] assessed the early-aged cracks effect function f(w) from regression analysis of field investigations at two different port wharves to predict a quantitative relationship between the chloride diffusion coefficient and crack width. The power function relationship for crack width wider than



Figure 1.10: Diffusion coefficient vs. crack width for mortar deformed under bending load [Sahmaran, 2007].



Figure 1.11: Partition hypothesis of chloride diffusion through cracked discs [Djerbi et al., 2008]

(0.1mm) is expressed as:

J

$$D_{cr} = f(w) \cdot D_a$$

$$f(w) = 31.61[|u|]^2 + 4.73[|u|] + 1, \quad ([|u|] \ge 0.1mm)$$
(1.19)

where D_a the apparent chloride diffusion coefficient. Then, this equation had been used to investigate the chloride behavior in cracked concrete.

Additionally, many studies reported that the growing effect of crack width on chloride ingress is imperceptible or constant if the value of crack opening is lower or upper a critical limits as stated in [Ismail et al., 2008, Djerbi et al., 2008, Jin et al., 2010, Jang et al., 2011]. They showed that only over a threshold crack width value, the diffusion coefficient starts to increase. The lower threshold width was

Reference	Lower limit (μm)	Upper limit (μm)
	(No effect of crack)	$(D_{cr} \text{ is constant})$
[Rodriguez and Hooton, 2003]	-	80-680
[Takewaka et al., 2003]	<50	>100
[Sahmaran, 2007]	<135	>135
[Ismail et al., 2008]	<30	>200
[Djerbi et al., 2008]	<30	>80
[Audenaert et al., 2009]	-	100-200
[Jin et al., 2010]	<30	>100
[Wang and Ueda, 2011a]	100	400
[Jang et al., 2011]	<55-80	>55-80
[Savija et al., 2013]	21	55

Table 1.1: Upper and lower limits of effective crack widths. [Gu et al., 2015] [Jin et al., 2010]

estimated by [Djerbi et al., 2008, Ismail et al., 2008, Jin et al., 2010] as $30\mu m$. Şahmaran [Sahmaran, 2007] found that for a crack width higher than $135\mu m$ the equivalent diffusion coefficient increases rapidly. However, the diffusion coefficient is still constant and crack wall can be considered as a chloride exposure surface once the crack width be greater than un upper value such as $200\mu m$ in [Ismail et al., 2008], $80\mu m$ in [Djerbi et al., 2008] and $100\mu m$ in [Jin et al., 2010]. More experimental results can be summarized in table 1.1.

The major difficulty facing the experimental investigation of the coupling is the complexity of the experimental setup implementation, a lot of time is demanded and the independent influences of certain parameters can not be identified. Hence, the importance of numerical studies as realistic as possible.

Regarding the numerical simulation of this coupling, some studies are particularly relevant. In Jin et al. [Jin et al.,], the authors integrate the diffusion coefficient through the crack obtained in Djerbi et al. [Djerbi et al., 2008] in their numerical model of diffusion based on the Finite Element method. Jin et al. [Jin et al.,] compare their numerical results with the experimental ones of Ismail et al. [Ismail et al., 2008]. Note that in the work of Jin et al. [Jin et al.,], only artificial crack is considered.

Wang and Ueda [Wang and Ueda, 2011a] consider a lattice-type model (see Grassl [Grassl, 2009] for details) as numerical model of diffusion. First Wang and Ueda [Wang and Ueda, 2011a] performed the trial and error method to fit in the experimental results of Ismail et al. [Ismail et al., 2008] again and find an equivalent diffusion coefficient. Then Wang and Ueda [Wang and Ueda, 2011a] consider a mesoscale lattice-type model with three phases (ITZ, aggregates, mortar) in order to gauge the influence of the ITZ and the aggregates on the depth of chloride penetration. The performance of this mesoscale model is assessed with the experimental results of Rodriguez et al. [Rodriguez and Hooton, 2003]. Here again in Wang and Ueda [Wang and Ueda, 2011a], only artificial crack is taken into account.

In Šavija et al. ([Savija et al., 2013] and [Savija et al., 2014]), the authors propose a numerical coupling between two models: for mechanical and diffusion simulations respectively, the Delft lattice model (see [Schlangen, 1993] for details) and the transport lattice model (see [Savija et al., 2013] for details). Contrary to the prior numerical works, mechanical simulations are performed leading to realisitic cracks. In Šavija et al. [Savija et al., 2014], according to findings of Yoon et al. [Yoon et al., 2007], the authors impose a chloride concentration only on cracks wider than 12 μ m obtained after the numerical simulation of a splitting test. In Šavija et al. [Savija et al., 2013], the authors assess their numerical model with (1) the experimental results of Şahmaran [Sahmaran, 2007] and Ismail et al. [Ismail et al., 2008] for cracked mortar samples by considering D_{cr} of Djerbi et al. [Djerbi et al., 2008], (2) with the experimental results of Ismail et al. [Ismail et al., 2004a] for cracked bricks samples – trial and error method to obtain the equivalent diffusion coefficient – in the context of artificial cracks.

1.5 Numerical modelling of heterogeneity of materials

Yet, in many applications, macroscopic properties -such as effective diffusivity- are only required as material constants in continuum theories. They can be gathered from subscale properties of material. Although, many studies usually treat concrete as a homogeneous material and estimate the diffusivity by fitting the experimental measurements of chloride concentration to evaluate the apparent diffusivity or the so-called "effective diffusion coefficient" neglecting the effects of individual phase properties [Xiao et al., 2012]. Concrete as a construction material is composed generally -at meso scale for instant- of three principal components: aggregate and cement paste and the interfacial transition zone. Each of them has a relevant properties concerning the mechanical and transport properties. Therefore, it is more successful to consider the individual properties in numerical simulation for better understanding of the mechanical and transport behavior of concrete as whole.

Other studies proposed multiphase analytical models to evaluate the effective

diffusion coefficient considering the individual transport properties and their volume fraction as in Hobbs [Hobbs, 1999] and Šavija et al. [Savija et al., 2013]. In [Hobbs, 1999], the author has presented an equation to estimate diffusion coefficient of concrete (D_c) as a function of diffusion coefficients of aggregate and cement paste as well as the aggregate volume fraction such as:

$$D_c = \frac{[(D_a - D_p)V_a + (D_p + D_a)]D_p}{[(D_p + D_a) + (D_p - D_a)V_a]}$$
(1.20)

where D_a and D_p are the chloride diffusivity of aggregate and cement paste respectively [Hobbs, 1999]. V_a represents the aggregate volume fraction. Šavija [Savija et al., 2013] used a similar approach that developed by Caré and Harvé (2014) [Caré and Hervé, 2004] to represent (D_{eff}) considering concrete as a three phases composite (ITZ, aggregate and mortar) and assuming impermeable aggregate as:

$$\frac{D_{eff}}{D_m} = \frac{N}{D} \tag{1.21}$$

where:

$$N = 6D_M(1 - C_A)(C_A + C_I) + 2C_I(D_I - D_M)(1 + 2C_A + 2C_I)$$
$$D = 3D_M(2 + C_A)(C_A + C_I) + 2C_I(1 - C_A - C_I)(D_I - D_M)$$

Here, D_M , and D_I are the diffusion coefficient of the mortar and the interface phase, respectively. Also, C_A and C_I are volume fractions of the aggregate and the interface phases, respectively.

Several authors have been suggested a discrete numerical methods using a lattice-type model on the mesoscale level which has the advantage of a more precise representation of transport properties for each phase [Wang and Ueda, 2011a]. Wang and Ueda achieved a such mesoscale lattice-type model with three phases(ITZ, mortar, aggregate) to estimate the influence of the ITZ and the aggregate on the chloride penetration depth [Wang and Ueda, 2011a], as shown in figure 1.12. In [Savija et al., 2013, Savija et al., 2014], the authors presented a



Figure 1.12: Construction of the lattice network model [Wang and Ueda, 2011a]

3D lattice model to predict chloride behavior in saturated and cracked concrete considering the transport (or mechanical) properties represented in each element. Here, the particle overlay procedure has been implemented to introduce the material heterogeneity (see Fig. 1.13). A computer generated material or material structure achieved by micro-CT scanning might be applied for this aim [Savija et al., 2013, Savija et al., 2014]. However, regarding this numerical simulation literature, the computation of macroscopic diffusivity tensors depending on its heterogeneous aspect is rarely performed especially when the heterogenities include the cracking state of concrete. From a civil engineering point of view, these macroscopic tensors are crucial regarding durability and service-life problems of civil engineering facilities. Therefore, it would be extremely helpful to predict the transport prop-



Figure 1.13: Two-dimensional particle overlay procedure [Savija et al., 2013]

erties of concrete based on its mesostructure. This is the main objective of my thesis.

To my knowledge, only the work of Nilenius et al. [Nilenius et al., 2015] provides such macroscopic information. Nilenius et al. implemented the concept of a statistical volume element (SVE) to generate a 3D model focusing on the mesoscale of concrete with consideration of cement paste, aggregates and ITZ [Nilenius et al., 2015].

1.6 Conclusion

In this chapter, a simple review of corrosion process of reinforcing bars of concrete, its causes and evolution stages were introduced to understand its effect on concrete deterioration and its service life. The chloride ingress is the main cause induce the reinforcement corrosion, so we presented the different mechanisms controlling chloride ions movement through cement-based materials such as diffusion, migration, convection and the chloride binding effects.

The experimental and numerical mechanical-transport coupling works were summarized and they show up the important and crucial role of cracks for increasing the acceleration of chloride penetration by forming a privileged path favoring the transport of chloride ions and consequently speed up the corrosion process. Regarding this numerical simulation literature, it would be intensely useful to estimate the transport properties of concrete at macroscale basing on its mesostructure.

As a result, a method is proposed here which has been found on an upscaling process in order to provide macroscopic diffusivity tensors accounting for the heterogeneous mesoscale structure of concrete with or without the contribution of chloride binding, and then correlate to the crack pattern and to the crack width values of numerically-induced cracks. In this context, important features of the cracking process are taken into account in the macroscopic diffusivity: (1) the evolution from diffuse cracks in the bulk to localized macro-crack(s), (2) the tortuosity of the crack pattern and (3) the induced-anisotropy.

In the following chapter, we will present the Finite Element formulation for the numerical modelling of chloride ions diffusion accounting for the different transport governing mechanisms as well as the chloride binding capacity in meso-scale concrete.

Chapter 2

Finite Element Modeling for Chloride Ions Transport Problem in Cement-Based Material

2.1 Introduction

Recently, a number of researches have been conducted on the modeling of ion transport mechanisms in concrete, Specifically in the case of chlorides, provided its effect on the corrosion of reinforcements mainly in marine environments or as a consequence of using salts as the deicing agent on bridges and roads.

The developed models take into account the material's parameters, environmental conditions and also regard to the different phenomenon that govern transport process like: diffusion, migration, capillarity suction and chloride binding. Various approaches have been used to model such process regarding the complexity of the transport mechanisms associated and material variation. One of the most commonly used method is the Finite Element Method.

In this chapter, we present the FE formulation for the numerical modelling of chloride ions diffusion accounting for different governing mechanism in meso-scale concrete. A review of chloride ions transport equations is briefly presented in section (2.2) to recall the governing equations used in the proposed numerical model.

The basic concepts of the FE method are used in section (2.3) to carry out a one dimensional solution of a transport problem to better understanding the formulation and the implementation of the FE solution code. Concrete is considered as a homogeneous material and diffusion is associated with some of other mechanisms such as migration and chloride binding.

While, in section (2.4) of this chapter, we turn to the Embedded Finite Element Method (E-FEM) in order to take into account the heterogeneity of concrete at the meso-scale where concrete is represented as aggregates embedded into a mortar matrix. We investigate through 1D simulations the influence of the microstructure properties and the chloride binding capacity on the chloride concentration profiles.

Comparison with analytic solutions are performed to show the accuracy of the proposed numerical models. We show that taking explicitly into account the diffusion coefficients of the microstructure and the chloride binding capacity are important to consider for a better prediction of the time span until corrosion initiation.

2.2 Review of chloride ions transport equations

As it is mentioned in chapter one, different mechanisms can control the chloride ions transport into concrete such as diffusion, permeation, migration and convection (or capillary suction) according to the surrounding conditions. The primary mechanism governing ions transport system in saturated porous media is diffusion which occurs due to a difference in concentration between the two sides of the material. Its mathematical expression are developed from the mass balance, energy conservation and constitutive equations where the concentration is the key variable.

In this case and neglecting any other transport mechanism, Ficks second law is obtained such as (see chapter one):

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \tag{2.1}$$

However, in the case of ionic species -due to the own electric charge or due to an external electrical field- the diffusion is governed by another mechanism and accordingly a different expression. The second modified Fick's law Eq. (2.2) is applicable for the molecular transport of each ionic species involved in the pore solution within the porous material in a one-dimensional fully saturated material [Rubinstein, 1990, Nilsson et al., 1996] such as:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - V D \frac{\partial c}{\partial x}$$
(2.2)

Further, if there is a physical or chemical interaction between chloride ions and the solid hydrates, D in Eq.(2.1) may be replaced by the effective diffusion coefficient

 $\left[D/(1+\frac{\partial c_b}{\partial c_f})\right]$ which represent the binding capacity. c_b is the concentration of bounded chloride ions and c_f is the concentration of free chloride ions. The relation between the bound and free chloride concentration can be mathematically described as "binding isotherm". Three well-known isotherms are used:

Linear isotherm: $c_b = \alpha c_f$

Langmuir isotherm: $c_b = \frac{\alpha_L c_f}{1 + \beta c_f}$

Freundlich isotherm: $\alpha_F c_f^{\beta_F}$

where α and β are experimentally founded parameters depending on the composition and properties of concrete. Although the chloride binding is not an instantaneous process and should take into account other factors such as *PH* of the solution and temperature, the binding rate is assumed to be instantaneous and has a perfect reversible equilibrium between free and bound chloride as in [Barbarulo et al., 2000].

2.3 Modeling of transport equation in homogeneous medium

It is evident that it does not exist an analytical solution to predict the exact data of chloride distribution when considering concrete as a multiphase domain or by achieving a three-dimensional simulation due to the variety of boundary conditions and material properties. Then, the best way to get approximate distribution is still the numerical methods like Finite Element formulation. The main goal in this section is to solve the pre-mentioned transport partial differential equations through Finite Element method to predict the chloride concentration c in time t and space C.

The mathematical models concerning chloride ions transport in concrete-like materials which were presented in the previous section introduce general types of differential equations depending on the mechanism that governs ions movement. In saturated state of concrete, the transport can be controlled by diffusion associated with chloride binding isotherm expressed by the second Fick's law or diffusion coupled by migration expressed by the modified Fick's second law.

However, although the intended characteristic of materials in this PhD thesis is to consider concrete as heterogeneous and cracked medium, it is important to show the Finite Element formulation for the simplest case assuming that concrete is homogeneous and sound (not cracked) material. This is owing to the need for understanding the Finite Element code FEAP with which solutions have been obtained, and also for achieving some model verifications with analytical solutions.

2.3.1 FEM formulation for the governing equations

As a general form, Eq. (2.2) is solved hereafter in order to get the concentration of chloride in time and space. Herein, the variational formulation of the relevant equations is derived. The boundary conditions assign the concentration c at the
Dirichlet boundary and flux J_c at the Neumann boundary:

$$c = c_o \quad on \quad \Gamma_c \tag{2.3}$$

$$\boldsymbol{J_c} = -D\frac{\partial c}{\partial x} \quad on \quad \Gamma_q \tag{2.4}$$

For a one-dimensional bar element, Γ_c corresponds to x = 0 and Γ_q to x = L.

The effect of diffusion, migration and nonlinear binding isotherm can be assembled in one equation to get a general finite element formulation. Then one gets:

$$div(\boldsymbol{J_c}) + \frac{\partial c}{\partial t} + VD\frac{\partial c}{\partial x} + \frac{\partial c_b}{\partial t} = 0$$
(2.5)

where c_b is the bounded chloride and Langmuir isotherm are chosen to define the relation between c_f and c_b .

2.3.2 Weak form of the problem

The last equation of chloride ions transport (2.5) is the strong form of the problem. In order to get the weak form, we choose a test function called w in the space C such as:

$$c = \{w(x) : \Omega \to \Re | w \in H^1, w = 0 \text{ on } \Gamma_c\}$$

$$\int_{\Omega} w(div(J_c) + \frac{\partial c}{\partial t} + V\frac{\partial c}{\partial x} + \frac{\partial c_b}{\partial t})d\Omega = 0$$
(2.6)

Expanding the former equation to get:

$$\int_{\Omega} w \left[-D\frac{\partial^2 c}{\partial x^2} \right] d\Omega + \int_{\Omega} w \left[\frac{\partial c}{\partial t} \right] d\Omega + V \int_{\Omega} w \frac{\partial c}{\partial x} d\Omega + \int_{\Omega} w \frac{\partial c_b}{\partial t} d\Omega = 0$$
(2.7)

By performing an integration by parts on the firs left side, the second-order derivative are replaced by first-order terms:

$$\left[-wD\frac{\partial c}{\partial x}\right]_{S} + D\int_{\Omega}\frac{\partial w}{\partial x}\frac{\partial c}{\partial x}d\Omega + \int_{\Omega}w\left[\frac{\partial C}{\partial t}\right]d\Omega + V\int_{\Omega}w\frac{\partial c}{\partial x}d\Omega + \int_{\Omega}w\frac{\partial c_{b}}{\partial t}d\Omega = 0 \quad (2.8)$$

Here, S in the first term denotes the element boundary such as $S = \Gamma_c \bigcup \Gamma_q$ where $\omega = 0$ on Γ_q has been used, then:

$$\left[-wD\frac{\partial c}{\partial x}\right]_{\Gamma_c} + D\int_{\Omega}\frac{\partial w}{\partial x}\frac{\partial c}{\partial x}d\Omega + \int_{\Omega}w\left[\frac{\partial c}{\partial t}\right]d\Omega + V\int_{\Omega}w\frac{\partial c}{\partial x}d\Omega + \int_{\Omega}w\frac{\partial c_b}{\partial t}d\Omega = 0 \quad (2.9)$$

Also,

$$q = -D\frac{\partial c}{\partial x} \text{ on } \Gamma_c \tag{2.10}$$

then,

$$D\int_{\Omega} \frac{\partial w}{\partial x} \frac{\partial c}{\partial x} d\Omega + \int_{\Omega} w [\frac{\partial c}{\partial t}] d\Omega + V \int_{\Omega} w \frac{\partial c}{\partial x} d\Omega + \int_{\Gamma_c} w q ds + \int_{\Omega} w \frac{\partial c_b}{\partial t} d\Omega = 0 \quad (2.11)$$

2.3.3 Spatial discretization of the problem

The next step is to discretize the domain Γ with an assemble of finite elements e. The unknown variables are expressed in terms of the nodal interpolation function N, also known as shape function, and the nodal unknown vectors.

$$c = N c^h$$

$$\omega = N \; \omega^h$$

where,

$$N = [N_1; N_2]$$
$$c^T = [c_1; c_2]^T$$

for a two-nodes one-dimensional bar element. According to the consistent formulation, the element nodal values of $\partial c/\partial t$ are expressed with vector $\{\dot{c}\}$ and the variation of $\partial c/\partial t$ throughout the element can similarly be expressed as:

$$\partial c/\partial t = N \{\dot{c}\}$$

Introducing these expressions (i.e. c, \dot{c} and ω) into the weak form equation (2.11), one gets:

$$\omega^{h,T} \left\{ \int_{\Omega} N^T D N d\Omega \ c + \int_{\Omega} B^T D B d\Omega \dot{c} + V \int_{\Omega} N^T B d\Omega c + \int_{\Gamma_q} N^T q dS + \int_{\Omega} N^T \frac{\partial c_b}{\partial t} d\Omega \right\} = 0$$

$$(2.12)$$

The above equation can be expressed in matrix form as:

$$\mathcal{K}c^{h} + \mathcal{M}\dot{c}^{h} + \mathcal{H}c^{h} + \int_{\Gamma_{q}} N^{T}qdS + \int_{\Omega} N^{T}\frac{\partial c_{b}}{\partial t}d\Omega = 0$$
(2.13)

where,

$$\mathcal{K} = \int_{\Omega} N^T D N d\Omega$$
 the diffusion matrix
 $\mathcal{M} = \int_{\Omega} B^T D B d\Omega$ the mass matrix

$$\mathcal{H} = V \int_{\Omega} N^T B d\Omega$$
 the convection matrix.

Note that we have:

$$N^{T} = [1 - x/l; x/l]^{T}$$

 $B^{T} = [-1/l; 1/l]^{T}$

B is a vector contains the derivatives of the shape functions for bar element of two nodes N_1 and N_2 (see Fig. (2.1)).



Figure 2.1: Shape functions N1 (left) and N2 (right)

Introducing these functions into the problem (2.13), one get:

$$\mathcal{K} = \frac{AD}{l} \begin{bmatrix} 1 & -1 \\ -1 & 1 \end{bmatrix}$$
(2.14)

$$\mathcal{M} = \frac{Al}{6} \begin{vmatrix} 2 & 1 \\ 1 & 2 \end{vmatrix}$$
(2.15)

$$\mathcal{H} = \frac{VAD}{2} \begin{bmatrix} -1 & 1\\ -1 & 1 \end{bmatrix}$$
(2.16)

where A the area of the element. We define the external and internal forces as

follows:

$$f^{ext} = \int_{\Gamma_q} N^T q dS$$

$$f^{int} = \mathcal{K}c^h + \mathcal{M}\dot{c}^h + \mathcal{H}c^h + \int_{\Omega} N^T \frac{\partial c_b}{\partial t} d\Omega$$
(2.17)

The goal is to minimize the residual R_1 at each time step t:

$$R_1(c) = f^{ext} - f^{int}$$

2.3.4 Discretization in time of problem

The time-dependent differential terms in Eq. (2.11) have to be discretized in time by applying the finite difference method as:

$$(\bullet) = \frac{(\bullet)|_{n+1} - (\bullet)|_n}{\Delta t} = \frac{\Delta(\bullet)}{\Delta t}$$
$$\mathcal{K}c^h + \mathcal{M}\frac{\Delta(c^h)}{\Delta t} + \mathcal{H}c^h + \int_{\Gamma_q} N^T q dS + \int_{\Omega} N^T \frac{\partial c_b}{\partial t} d\Omega = 0$$

Here, the last temporal variation term of chloride binding where the Langmuir nonlinear isotherm has been used as:

$$c_b = \frac{\alpha_L N c_f}{1 + \beta N c_f}$$
, and

$$\frac{\partial c_b}{\partial t} = \frac{c_b^h|_{n+1} - c_b^h|_n}{\Delta t} = \frac{\Delta(c_b^h)}{\Delta t}$$

then,

$$\mathcal{K}c^{h} + \frac{\mathcal{M}}{\Delta t}\Delta c^{h} + \mathcal{H}c^{h} + \int_{\Omega} N^{T} \frac{\Delta(c_{b}^{h})}{\Delta t} d\Omega + \int_{\Gamma_{q}} N^{T} q dS = 0$$

2.3.5 Linearization of the problem

In order to solve the resulting nonlinear equation, it is necessary to accomplish a consistent linearization to obtain a linear system. We will now linearize discrete weak form Eq. (2.13) with respect to the value of chloride ions concentration c by carrying out k iterations at each time step n until achieving the convergence of the problem.

Let R_1 be an equilibrium equation that depends on \mathcal{X}_i variables, the linearization of R_1 with respect to \mathcal{X}_i is:

$$\mathcal{L}\left[R_1|_{n+1}^{k+1}\right] \simeq R_1|_{n+1}^k + \frac{\partial R_1}{\partial \mathcal{X}_i}|_{n+1}^k \cdot \Delta \mathcal{X}_i|_{n+1}^{k+1} = 0$$
(2.18)

$$R_1|_{n+1}^k = \left[\mathcal{K}c^h + \frac{\mathcal{M}}{\Delta t}\Delta c^h + \mathcal{H}c^h + \int_{\Omega} N^T \frac{\Delta(c_b^h)}{\Delta t} d\Omega + \int_{\Gamma_q} N^T q dS\right]\Big|_{n+1}^k$$
(2.19)

$$\Delta c_b^h = \begin{cases} = c_b^h |_{n+1}^k - c_b^h|_n \\ \\ = \frac{\alpha_L N c_f^h |_{n+1}^k}{1 + \beta N c_f^h |_{n+1}^h} - \frac{\alpha_L N c_f^h |_n}{1 + \beta N c_f^h |_n} \end{cases}$$
(2.20)

also,

$$\left. \frac{\partial c_b}{\partial c_f} \right|_{n+1}^k = \frac{\alpha_L N}{(1 + \beta N c_f^h |_{n+1}^k)^2}, \text{according to the quotient rule}$$

then,

 \Rightarrow

 \Rightarrow

$$\mathcal{K}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \frac{\mathcal{M}}{\Delta t}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \mathcal{H}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \int_{\Omega} \frac{N^{T}}{\Delta t} \left(\frac{\partial c_{b}}{\partial c_{f}} \Big|_{n+1}^{k} - \frac{\partial c_{b}}{\partial c_{f}} \Big|_{n} \right) \Delta c|_{n+1}^{k+1} d\Omega$$

$$(2.21)$$

$$\mathcal{K}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \frac{\mathcal{M}}{\Delta t}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \mathcal{H}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \int_{\Omega} \frac{N^{T}}{\Delta t} \frac{\alpha_{L}N}{(1+\beta N c_{f}^{h}|_{n+1}^{k})^{2}} \Delta c|_{n+1}^{k+1} d\Omega$$
(2.22)

$$\mathcal{K}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \frac{\mathcal{M}}{\Delta t}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \mathcal{H}|_{n+1}^{k}\Delta c|_{n+1}^{k+1} + \int_{\Omega} \frac{N^{T}.N}{\Delta t} \frac{\alpha_{L} d\Omega}{(1+\beta N c_{f}^{h}|_{n+1}^{k})^{2}} \Delta c|_{n+1}^{k+1}$$
(2.23)

Considering \mathcal{M}_L is equal to $\int_{\Omega} \frac{N^T N}{\Delta t} \frac{\alpha_L d\Omega}{(1+\beta N c_f^h|_{h+1}^k)^2}$, the last equation will be:

$$\left[\mathcal{K} + \frac{\mathcal{M}}{\Delta t} + \frac{\mathcal{M}_L}{\Delta t} + \mathcal{H}\right] \Delta c|_{n+1}^{k+1} = -\frac{\partial R_1}{\partial c_i}|_{n+1}^k \tag{2.24}$$

To numerically evaluate these terms, two gauss points are applied to each element, and numerical integration is implemented. In this way, the transformation of global coordinate x_i to local coordinate (ζ_i) has been used considering the Gauss-Legender formula (with their corresponding weight W_i at points of numerical integration). Once Δc is known and converged for a given (k) called (k_{cv}) , we can update the values for the time step n + 1 to find the values of c:

$$c_{n+1} = c_n + \Delta c^{(k_{cv})}$$

The next section will be the opportunity to validate the numerical model behavior by comparing model output to corresponding output obtained from the analytical solution.

2.3.6 Numerical results and validation of model

Implementation of the finite element code has been performed using a user subroutine through a computer analysis system FEAP [Taylor, 2008]. The model is validated here using the exact solution of one-dimensional transient chloride transport problem. As shown previously, Eq. (2.5) is considered as a general representation of transport problem in saturated concrete-like materials to describe three phenomena: diffusion, migration and chloride binding. Therefore, it is important to use the analytical solution corresponding to each transport equation that contains one or more phenomenon like diffusion with/without migration or chloride binding as mentioned in chapter one.

In order to illustrate the performance of our model, a simple numerical example considering a homogeneous bar of 100 mm long and 1 mm^2 area is simulated as a one-dimension flow problem. For a semi-infinite medium, the boundary and initial conditions are:

$$\begin{cases} c = c_0 \quad \text{for} \quad x = x_0 \quad \text{and} \quad t > 0 \\ c = 0 \quad \text{for} \quad x > 0 \quad \text{and} \quad t = 0 \end{cases}$$
(2.25)

The closed-solution of the problem governed only by diffusion can be obtained

-as defined in chapter one-:

$$c(x,t) = c_0 \left(1 - erf\left(\frac{x}{2\sqrt{Dt}}\right) \right)$$
(2.26)

where erf() is the standard error function.

The above equation and the other exact solution -used here- are valid only when the coefficient of diffusion D and the surface chloride concentration c_0 are assumed to be constant (except when considering chloride binding effect). Moreover, if migration is considered to provoke chloride ions transport in addition to diffusion mechanism, the next exact solution is used to compare the numerical results such as:

$$c(x,t) = \frac{c_0}{2} \left(e^{Vx} erfc\left(\frac{x+VDt}{2\sqrt{Dt}}\right) + erfc\left(\frac{x-VDt}{2\sqrt{Dt}}\right) \right)$$
(2.27)

for a semi-infinite medium (cf. chapter one), where erfc is the complementary error function defined as:

$$erfc(x) = 1 - erf(x)$$

Additionally, if the contribution of chloride binding is regarded, a comparison with modified Crank's solution of Fick's law is implemented in two steps. Firstly, by calculating the profile of chloride concentration without binding contribution and using the nonlinear Langmuir isotherm next to modify the coefficient of diffusion as in Eq. (2.28).

$$D_L = \frac{D_c}{1 + \frac{\alpha_L}{[1 + \beta_L C(x,t)]^2}}$$
(2.28)

where D_L is the modified diffusion coefficient computed at each node.

After that, the solution is updated at each node to find the free chloride concentration c_f using the updated values of diffusion coefficient as in Eq. (2.29).

$$c(x,t) = c_0 \left(1 - erf\left(\frac{x}{2\sqrt{D_L t}}\right) \right)$$
(2.29)

To examine the numerical model, a bar of 100 mm is discretized with 100 twonode elements. Diffusion coefficient D_c , electrostatic potential gradient E and temperature T are summarized in Table (2.1) as used in [Savija et al., 2014].

$D_c \ (m^2/s)$	$E(V.m^{-1})$	T(K)	α_L	β_L
2.78×10^{-11}	1000	293	1.67	4.08

 Table 2.1: Values for the numerical simulations : chloride diffusion with chloride binding.

Values of Langmuir isotherm constants α_L and β_L were taken according to regression analysis of the experimental data of [Sergi et al., 1992a], as shown in Table (2.1).

A chloride surface concentration c_0 equals to $10^{-2}kg.m^{-3}$ is imposed at x = 0 at the left end of the bar. Figure 2.2 plots the resulting chloride concentration profiles of finite element solution and exact solution of Eq. (2.1). It is notable that the finite element simulation results compare very well with analytical solution.

Figures (2.3) and (2.4) also compare the numerical results with the analytical solution for Eq. (2.2) considering diffusion, migration and chloride binding. These figures exhibit good agreement between numerical results and results from their suitable analytic solution again.



Figure 2.2: Finite element result and analytical solution considering diffusion



Figure 2.3: Finite element result and analytical solution considering diffusion and migration

2.4 Modeling of transport equation considering

heterogeneous characteristics of concrete

One of the objectives of this chapter is to present a numerical model capable of integrating material's variation with the physical phenomena at the mesoscale which plays a significant role in the behavior of concrete.



Figure 2.4: Finite element result and analytical solution considering diffusion, migration and nonlinear isotherm

For this reason, we follow a special kinematic enhancement leading to finite strain jump (discontinuity) at the interface in a typical truss element with different diffusion properties which denominated as "Weak discontinuity" [Benkemoun et al., 2012]. Therefore, certain elements are found to be split into two parts each with different material properties (see Fig. (2.5)).



Figure 2.5: Bar element split into two parts

The Embedded Finite Element Method (E-FEM, [Armero and Garikipati, 1996, Oliver, 1996, Borja, 2000, Benkemoun et al., 2010]) will be used here after to retain all the advantages of the finite element approach (applicability to different constitutive laws, robustness, compatibility with homogenization techniques, ...) while considerably easing the meshing step of physical discontinuities such as material interfaces.

2.4.1 Governing equation : diffusion accounting for chloride binding

For a saturated concrete where diffusion process controls transport mechanism of chloride ions and taking into account the chloride binding capacity, the governing equation of chloride diffusion can be described as:

$$\frac{\partial c_t}{\partial t} + \nabla \cdot (\boldsymbol{J_{c_f}}) = 0 \tag{2.30}$$

where $c_t = c_f + c_b$ is the total chloride concentration [mol.m⁻³], representing the sum of free chloride concentration c_f and bound chloride concentration c_b . J_{C_f} [J.m⁻².s⁻¹] is the flux accounting for chloride ions transport by diffusion. It is such as :

$$\boldsymbol{J_{c_f}} = -D\nabla(c_f) \tag{2.31}$$

where D is the diffusion coefficient $[m^2.s^{-1}]$.

Substituting $c_t = c_f + c_b$ and Eq. (2.31) in Eq. (2.30) yield :

$$\frac{\partial c_f}{\partial t} + \frac{\partial c_b}{\partial t} - \nabla \cdot (D\nabla(c_f)) = 0$$
(2.32)

Assuming a one dimensional problem through each lattice element, Eq. (2.32) becomes :

$$\frac{\partial c_f}{\partial t} + \frac{\partial c_b}{\partial t} - D \frac{\partial^2 c_f}{\partial x^2} = 0$$
(2.33)

where D is assumed to be constant.

In this work, the *Langmuir* isotherm model is used to demonstrate the practicability of the Embedded Finite Element method for chloride ions diffusion modeling with binding isotherm.

Consequently, $\frac{\partial c_b}{\partial t}$ can be written such as :

$$\frac{\partial c_b}{\partial t} = \frac{\partial c_b}{\partial c_f} \frac{\partial c_f}{\partial t},\tag{2.34}$$

2.4.2 Embedded Finite Element formulation

The discrete form of Eq. (2.33) is obtained by employing the Galerkin approximation. This approximation leads to (cf. [Benkemoun et al., 2017a]):

$$\int_{\Omega} \delta c_f \frac{\partial c_f}{\partial t} d\Omega + \int_{\Omega} \delta c_f \frac{\partial c_b}{\partial t} d\Omega + \int_{\Omega} \frac{\partial \delta c_f}{\partial x} D \frac{\partial c_f}{\partial x} d\Omega - \int_{\partial \Omega_{J_{c_f}}} \delta c_f \bar{J}_{c_f} d\partial \Omega = 0 \quad (2.35)$$

where δc_f is the virtual free chloride concentration in the space \mathcal{C} such as $\mathcal{C} = \{\delta c_f(x) : \Omega \mapsto \mathbb{R} \mid \delta c_f \in H^1, \delta c_f = 0 \text{ on } \partial \Omega_{c_f} \}$. We note $\partial \Omega_{c_f}$ the part of the boundary where the free chloride concentration is imposed and $\partial \Omega_{J_{c_f}}$ where the flux \overline{J}_{c_f} is imposed.

As it is earlier stated in section (2.4) for the finite elements that split into two parts Ω^{\oplus} and Ω^{\oplus} , each part having different diffusivity properties, $D^{\oplus}, \alpha_L^{\oplus}, \beta_L^{\oplus}$ and $D^{\oplus}, \alpha_L^{\oplus}, \beta_L^{\oplus}$. We note $D^{\oplus/\oplus}, \alpha_L^{\oplus/\oplus}, \beta_L^{\oplus/\oplus}$ the diffusivity coefficient and the chloride binding parameters in $\Omega^{\oplus/\oplus}$. In order to take into account this material interface in these elements, the finite element space is enriched through a weak discontinuity, located at the interface of both materials. As performed in [Ibrahimbegovic and Wilson, 1991a] this weak discontinuity is introduced in the kinematics of both the real and virtual free chloride concentration fields such as c_f and δc_f are:

$$c_f = \underbrace{\bar{c}_f}_{\text{regular}} + \underbrace{\bar{\bar{c}}_f}_{\text{enriched}}$$
(2.36)

$$\delta c_f = \underbrace{\delta \bar{c}_f}_{\text{regular}} + \underbrace{\delta \bar{\bar{c}}_f}_{\text{enriched}}$$
(2.37)

As in [Simo and Rifai, 1990], we refer to \overline{c}_f and $\delta \overline{c}_f$ as the enriched parts of the free chloride concentration fields. The notation ($\overline{\bullet}$) refers to an enrichment based upon a weak discontinuity.

Having the form of c_f and δc_f , we now turn to the discretization of these fields such as [Benkemoun et al., 2015]:

$$c_f = \mathbf{N}\bar{\mathbf{c}}_f^h + M^{\oplus/\ominus}\bar{\bar{c}}_f^h \tag{2.38}$$

$$\delta c_f = \mathbf{N} \delta \bar{\mathbf{c}}_f^h + M^{\oplus/\ominus} \delta \bar{\bar{c}}_f^h \tag{2.39}$$

where N is a row vector containing the classical shape functions for a bar element. \bar{c}_f^h and $\delta \bar{c}_f^h$ are column vector containing the real and virtual free chloride concentration nodal unknowns respectively. The mathematical form of $M^{\oplus/\ominus}$ represents a scalar function. Fig. (2.6) plots $M^{\oplus/\ominus}$ in the 1D case knowing that l^e the length of the element and θl^e the position of the jump of the diffusivity properties. The form of N and $M^{\oplus/\ominus}$ are known as:

$$\begin{split} N^T &= [1 - x/l; x/l]^T \\ M^{\ominus} &= -x/(\theta l), \quad \text{if } x \in [0, \theta l] \\ M^{\oplus} &= x/(l(1 - \theta)) - 1/(1 - \theta), \quad \text{if } x \in [\theta l, l] \end{split}$$

Note that the function $M^{\oplus/\ominus}$ is also selected in [Moës et al., 2003] for the X-FEM. They prove that the choice of this function gives convergence rate very close to the optimal finite element convergence.



Figure 2.6: Enriched function $M^{\oplus/\Theta}$ for a typical split element [Benkemoun et al., 2017a].

Including the pre-mentioned expressions 2.36 and 2.37 in Eq. (2.35), the derivatives of c_f and δc_f in relation with x are needed. Considering Eq. (2.38) and (2.39), the derivatives are such as :

$$\frac{\partial c_f}{\partial x} = \boldsymbol{B}\bar{\boldsymbol{c}}_f^h + G^{\oplus/\ominus}\bar{\bar{c}}_f^h \tag{2.40}$$

$$\frac{\partial \delta c_f}{\partial x} = \boldsymbol{B} \delta \bar{\boldsymbol{c}}_f^h + G^{\oplus/\ominus} \delta \bar{\bar{c}}_f^h \tag{2.41}$$

where \boldsymbol{B} is a row vector containing the classical shape functions derivatives for a

bar element. Considering the form of $M^{\oplus/\ominus}$ in Fig. (2.6), Fig. (2.7) plots $G^{\oplus/\ominus}$ in the 1D case knowing that l^e the length of the element and θl^e the position of the jump of the diffusivity properties. $G^{\oplus/\ominus}$ is a piecewise linear function over a typical split element capturing the jump of the diffusivity properties in the gradient of the free chloride concentration field. The form of **B** vector and $G^{\oplus/\ominus}$ function is known such as:

$$B^{T} = [-1/l; 1/l]^{T}$$

$$G^{\oplus} = -1/(\theta l), \quad \text{if } x \in [0, \theta l]$$

$$G^{\oplus} = 1/l(1-\theta), \quad \text{if } x \in [\theta l, l]$$



Figure 2.7: Enriched function $G^{\oplus/\ominus}$ for a typical split element [Benkemoun et al., 2017a].

By inserting Eq. (2.38), (2.39), (2.40) and (2.41) in (2.35) for any $\delta \bar{c}_f^h$ and $\delta \bar{c}_f^h$, we obtain the following:

$$\begin{cases} \mathcal{M}\dot{\bar{c}}_{f}^{h} + \mathcal{P}\dot{\bar{c}}_{f}^{h} + \mathcal{K}\bar{c}_{f}^{h} + \mathcal{F}\bar{\bar{c}}_{f}^{h} + \int_{\Omega} \mathbf{N}^{T}\frac{\partial c_{b}}{\partial t} = \mathbf{f}^{ext} \\ \mathcal{P}^{T}\dot{\bar{c}}_{f}^{h} + \mathcal{L}\dot{\bar{c}}_{f}^{h} + \mathcal{F}^{T}\bar{c}_{f}^{h} + \mathcal{H}\bar{\bar{c}}_{f}^{h} + \int_{\Omega} M^{\oplus/\ominus}\frac{\partial c_{b}}{\partial t} = 0 \end{cases}$$
(2.42)

where (\bullet) represents time derivatives and where the flux \bar{J}_{c_f} is applied only on the regular part of the free chloride concentration field.

We note

$$\mathcal{M} = \int_{\Omega} \mathbf{N}^{T} \mathbf{N} d\Omega, \qquad \mathcal{P} = \int_{\Omega} \mathbf{N}^{T} M^{\oplus/\ominus} d\Omega \qquad (2.43)$$

$$\mathcal{K} = \int_{\Omega} \boldsymbol{B}^{T} D \boldsymbol{B} d\Omega, \qquad \mathcal{F} = \int_{\Omega} \boldsymbol{B}^{T} D G^{\oplus/\ominus} d\Omega \qquad (2.44)$$

$$\mathcal{L} = \int_{\Omega} M^{\oplus/\ominus} M^{\oplus/\ominus} d\Omega, \qquad \mathcal{H} = \int_{\Omega} G^{\oplus/\ominus} DG^{\oplus/\ominus} d\Omega$$
(2.45)

$$\boldsymbol{f}^{ext} = \int_{\partial\Omega_{J_{c_f}}} \boldsymbol{N}^{\boldsymbol{T}} \bar{J}_{c_f} d\partial\Omega \qquad (2.46)$$

where $\mathcal{M}, \mathcal{P}, \mathcal{K}, \mathcal{F}, \mathcal{L}$ and \mathcal{H} are directly integrated to obtain:

$$\mathcal{M} = \frac{A^{e}l^{e}}{6} \begin{pmatrix} 2 & 1 \\ 1 & 2 \end{pmatrix}$$
$$\mathcal{P} = \frac{A^{e}l^{e}}{6} \begin{pmatrix} (\theta - 2) \\ -(\theta + 1) \end{pmatrix}$$
$$\mathcal{K} = \frac{A^{e}(D^{\ominus}\theta + D^{\oplus}(1 - \theta))}{l^{e}} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix}$$
$$\mathcal{F} = \frac{A^{e}(D^{\ominus} - D^{\oplus})}{l^{e}} \begin{pmatrix} 1 \\ -1 \end{pmatrix}$$
$$\mathcal{L} = \frac{A^{e}l^{e}}{3} \qquad (2.47)$$

$$\mathcal{H} = A^e \left[\frac{D^{\ominus}}{\theta l^e} + \frac{D^{\oplus}}{(1-\theta)l^e}\right]$$
(2.48)

We note A^e the section of a bar element.

In Eq. (2.42), $\frac{\partial c_b}{\partial t}$ is expressed as in Eq. (2.34) where $\frac{\partial c_b}{\partial c_f}$ represents the chloride binding capacity. By considering the *Langmuir* isotherm model in Eq. (2.34), $\frac{\partial c_b}{\partial c_f}$ can be rewritten such as:

$$\frac{\partial c_b}{\partial c_f} = \frac{\alpha_L}{(1 + \beta_L c_f)^2}.$$
(2.49)

Now, considering Eq. (2.34) with Eq. (2.49), the system (2.42) becomes :

$$\begin{cases} \mathcal{M}\dot{\bar{c}}_{f}^{h} + \mathcal{P}\dot{\bar{c}}_{f}^{h} + \mathcal{K}\bar{c}_{f}^{h} + \mathcal{F}\bar{\bar{c}}_{f}^{h} + \mathcal{M}_{\mathcal{L}}(c_{f})\dot{\bar{c}}_{f}^{h} + \mathcal{P}_{\mathcal{L}}(c_{f})\dot{\bar{c}}_{f}^{h} = \boldsymbol{f}^{ext} \\ \mathcal{P}^{T}\dot{\bar{c}}_{f}^{h} + \mathcal{L}\dot{\bar{c}}_{f}^{h} + \mathcal{F}^{T}\bar{c}_{f}^{h} + \mathcal{H}\bar{c}_{f}^{h} + \mathcal{P}_{\mathcal{L}}^{T}(c_{f})\dot{\bar{c}}_{f}^{h} + \mathcal{L}_{\mathcal{L}}(c_{f})\dot{\bar{c}}_{f}^{h} = 0 \end{cases}$$
(2.50)

We note

$$\mathcal{M}_{\mathcal{L}}(c_f) = \int_{\Omega} \mathbf{N}^T \frac{\alpha_L}{(1+\beta_L c_f)^2} \mathbf{N} d\Omega, \qquad \mathcal{P}_{\mathcal{L}}(c_f) = \int_{\Omega} \mathbf{N}^T \frac{\alpha_L}{(1+\beta_L c_f)^2} M^{\oplus/\Theta} d\Omega$$
(2.51)

$$\mathcal{L}_{\mathcal{L}}(c_f) = \int_{\Omega} M^{\oplus/\ominus} \frac{\alpha_L}{(1 + \beta_L c_f)^2} M^{\oplus/\ominus} d\Omega$$
(2.52)

The terms in $\mathcal{M}_{\mathcal{L}}(c_f)$, $\mathcal{P}_{\mathcal{L}}(c_f)$ and $\mathcal{L}_{\mathcal{L}}(c_f)$ are evaluated using the numerical integration at the Gauss points of each sub-domain Ω^{\oplus} and Ω^{\oplus} . For instance $\mathcal{M}_{\mathcal{L}}(c_f)$ is computed through the technology of isoparametric FE such as :

$$\mathcal{M}_{\mathcal{L}}(c_f) \simeq \mathbf{N}^{\mathbf{T}}(\xi^{\oplus}) \frac{\alpha_L^{\oplus}}{(1 + \beta_L^{\oplus} c_f(\xi^{\oplus}))^2} \mathbf{N}(\xi^{\oplus}) j^{\oplus} w^{\oplus} + \mathbf{N}^{\mathbf{T}}(\xi^{\ominus}) \frac{\alpha_L^{\ominus}}{(1 + \beta_L^{\oplus} c_f(\xi^{\ominus}))^2} \mathbf{N}(\xi^{\ominus}) j^{\oplus} w^{\ominus}$$

$$(2.53)$$

where $\xi^{\oplus(\ominus)}$, $j^{\oplus(\ominus)}$ and $w^{\oplus(\ominus)}$ are respectively the position of the Gauss points, the determinant of the Jacobian and the weight in $\Omega^{\oplus(\ominus)}$. We also note that different values for α_L and β_L can be considered in Ω^{\oplus} and Ω^{\ominus} .

$$\begin{pmatrix} \boldsymbol{f}_{n+1}^{int,(k)} = \left[\frac{\mathcal{M}}{\Delta t} + \frac{\mathcal{M}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\boldsymbol{c}}_{f}^{h}|_{n+1}^{(k)} + \mathcal{K} \bar{\boldsymbol{c}}_{f}^{h}|_{n+1}^{(k)} + \left[\frac{\mathcal{P}}{\Delta t} + \frac{\mathcal{P}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\bar{c}}_{f}^{h}|_{n+1}^{(k)} + \mathcal{F} \bar{\bar{c}}_{f}^{h}|_{n+1}^{(k)} \\ h_{n+1}^{(k)} = \left[\frac{\mathcal{P}^{T}}{\Delta t} + \frac{\mathcal{P}_{\mathcal{L}}^{T}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\boldsymbol{c}}_{f}^{h}|_{n+1}^{(k)} + \mathcal{F}^{T} \bar{\boldsymbol{c}}_{f}^{h}|_{n+1}^{(k)} + \left[\frac{\mathcal{L}}{\Delta t} + \frac{\mathcal{L}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\bar{c}}_{f}^{h}|_{n+1}^{(k)} + \mathcal{H} \bar{\bar{c}}_{f}^{h}|_{n+1}^{(k)} \\ (2.54) \end{cases}$$

In the transient equation (2.54), the Newmark time integration scheme has been considered for time dependent terms.

2.4.3 Linearization of the problem

Although several schemes are possible, here we consider obtaining the incremental updates of the dependent variables $\Delta \bar{c}_{f}^{h}|_{n+1}^{(k+1)}$ and $\Delta \bar{\bar{c}}_{f}^{h}|_{n+1}^{(k+1)}$ by linearizing $f_{n+1}^{int,(k)}$ and $h_{n+1}^{(k)}$ about the current state, defined by $\bar{c}_{f}^{h}|_{n+1}^{(k)}$ and $\bar{\bar{c}}_{f}^{h}|_{n+1}^{(k+1)}$ as in [Benkemoun et al., 2017a].

By using equation (2.18), the linearization of system (2.54) leads to :

$$\left\{ \begin{array}{l} -\boldsymbol{R}_{1n+1}^{(k)} = \left[\frac{\mathcal{M}}{\Delta t} + \mathcal{K} + \frac{\mathcal{M}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\boldsymbol{c}}_{f}^{h} \Big|_{n+1}^{(k+1)} + \left[\frac{\mathcal{P}}{\Delta t} + \mathcal{F} + \frac{\mathcal{P}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\bar{c}}_{f}^{h} \Big|_{n+1}^{(k+1)} \\ -\boldsymbol{R}_{2n+1}^{(k)} = \left[\frac{\mathcal{P}^{T}}{\Delta t} + \mathcal{F}^{T} + \frac{\mathcal{P}_{\mathcal{L}}^{T}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\boldsymbol{c}}_{f}^{h} \Big|_{n+1}^{(k+1)} + \left[\frac{\mathcal{L}}{\Delta t} + \mathcal{H} + \frac{\mathcal{L}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \Delta \bar{\bar{c}}_{f}^{h} \Big|_{n+1}^{(k+1)} \\ \end{array} \right.$$
(2.55)

System (2.55) represents the system to be solved in terms of $\Delta \bar{c}_{f}^{h}|_{n+1}^{(k+1)}$ and $\Delta \bar{c}_{f}^{h}|_{n+1}^{(k+1)}$. Hereafter, we present the numerical solving strategy for the resolution of (2.55).

2.4.4 Global solution strategy

Consider $(\bullet)|_{n+1}^{(k)}$ is presumed to be the value of the variable (\bullet) at the k-th iteration of an incremental time step $[t_n; t_{n+1}]$. The numerical solution strategy is attempted such as(cf. [Benkemoun et al., 2017a]):

i) Expressing the incremental variable $\Delta \bar{c}_{f}^{h|(k+1)}_{n+1}$ as a function of $\Delta \bar{c}_{f}^{h|(k+1)}_{n+1}$ by using the second part of system (2.55) such as:

$$\Delta \bar{c}_{f}^{h}|_{n+1}^{(k+1)} = -\boldsymbol{R}_{2n+1}^{(k)} \left[\left[\frac{\mathcal{L}}{\Delta t} + \mathcal{H} + \frac{\mathcal{L}_{\mathcal{L}}(c_{f})}{\Delta t} \right]_{n+1}^{(k)} \right]^{-1} - \left[\left[\frac{\mathcal{L}}{\Delta t} + \mathcal{H} + \frac{\mathcal{L}_{\mathcal{L}}(c_{f})}{\Delta t} \right]_{n+1}^{(k)} \right]^{-1} \left[\frac{\mathcal{P}^{T}}{\Delta t} + \mathcal{F}^{T} + \frac{\mathcal{P}_{\mathcal{L}}^{T}(c_{f})}{\Delta t} \right]_{n+1}^{(k)} \Delta \bar{c}_{f}^{h}|_{n+1}^{(k+1)}$$
(2.56)

ii) Combining (2.56) with the second part of system (2.55), $\Delta \bar{c}_{f|n+1}^{h|(k+1)}$ is obtained as the solution of:

$$\begin{bmatrix} \left[\frac{\mathcal{M}}{\Delta t} + \mathcal{K} + \frac{\mathcal{M}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} - \left[\frac{\mathcal{P}}{\Delta t} + \mathcal{F} + \frac{\mathcal{P}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \begin{bmatrix} \left[\frac{\mathcal{L}}{\Delta t} + \mathcal{H} + \frac{\mathcal{L}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \end{bmatrix}^{-1} \\ \begin{bmatrix} \frac{\mathcal{P}^{T}}{\Delta t} + \mathcal{F}^{T} + \frac{\mathcal{P}_{\mathcal{L}}^{T}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \\ = -\mathbf{R}_{1n+1}^{(k)} + \left[\frac{\mathcal{P}}{\Delta t} + \mathcal{F} + \frac{\mathcal{P}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \mathbf{R}_{2n+1}^{(k)} \left[\left[\frac{\mathcal{L}}{\Delta t} + \mathcal{H} + \frac{\mathcal{L}_{\mathcal{L}}(c_{f})}{\Delta t}\right]_{n+1}^{(k)} \right]^{-1}$$
(2.57)

iii) System (2.57) can be written in a compact form such as :

$$\hat{K}_{n+1}^{(k)} \Delta \bar{\boldsymbol{c}}_{f}^{h} \big|_{n+1}^{(k+1)} = \hat{F}_{n+1}^{(k)}$$
(2.58)

where

$$\hat{K}_{n+1}^{(k)} = \left[\left[\frac{\mathcal{M}}{\Delta t} + \mathcal{K} + \frac{\mathcal{M}_{\mathcal{L}}(c_f)}{\Delta t} \right]_{n+1}^{(k)} - \left[\frac{\mathcal{P}}{\Delta t} + \mathcal{F} + \frac{\mathcal{P}_{\mathcal{L}}(c_f)}{\Delta t} \right]_{n+1}^{(k)} \left[\left[\frac{\mathcal{L}}{\Delta t} + \mathcal{H} + \frac{\mathcal{L}_{\mathcal{L}}(c_f)}{\Delta t} \right]_{n+1}^{(k)} \right]_{n+1}^{-1} \right]_{n+1}^{-1} \left[\frac{\mathcal{P}^T}{\Delta t} + \mathcal{F}^T + \frac{\mathcal{P}_{\mathcal{L}}^T(c_f)}{\Delta t} \right]_{n+1}^{(k)} \right]$$
(2.59)

and

$$\hat{F}_{n+1}^{(k)} = -[\boldsymbol{f}_{n+1}^{int,(k)} - \boldsymbol{f}_{n+1}^{ext}] + [\frac{\mathcal{P}}{\Delta t} + \mathcal{F} + \frac{\mathcal{P}_{\mathcal{L}}(c_f)}{\Delta t}]_{n+1}^{(k)} h_{n+1}^{(k)} \left[[\frac{\mathcal{L}}{\Delta t} + \mathcal{H} + \frac{\mathcal{L}_{\mathcal{L}}(c_f)}{\Delta t}]_{n+1}^{(k)} \right]^{-1}$$
(2.60)

iv) Once $\Delta \bar{c}_{f}^{h}|_{n+1}^{(k+1)}$ is known, $\Delta \bar{\bar{c}}_{f}^{h}|_{n+1}^{(k+1)}$ is computed through (2.56).

v) Check convergence, when those values have converged for a given (k) called (k_{cv}) , then $(\bullet)|_{n+1}^{(k_{cv})}$ is the solution and we can update the values for the time step n+1:

$$\bar{c}_{f}^{h}|_{n+1} = \bar{c}_{f}^{h}|_{n} + \Delta \bar{c}_{f}^{h}|_{n+1}^{(k_{cv})}$$
(2.61)

$$\bar{\bar{c}}_{f}^{h}|_{n+1} = \bar{\bar{c}}_{f}^{h}|_{n} + \Delta \bar{\bar{c}}_{f}^{h}|_{n+1}^{(k_{cv})}$$
(2.62)

2.4.5 Numerical simulations for 1D case and comparison with analytical solutions

Here we show numerical simulations which investigate the accuracy of the Embedded Finite Element method for the numerical modeling of chloride ions diffusion in a two-phase material accounting for chloride binding capacity. As first step, the case without chloride binding is also studied. For one-dimensional simulations and semi-infinite two-phase domain, we consider a bar of length 200 mm and area 1 mm^2 for all the simulations. This bar is split into two sub-domains Ω^{\ominus} and Ω^{\oplus} at x = 28.5 mm and is discretized with 200 two-node bar element elements connected in series and aligned in the *x*-direction. The finite element discretization is illustrated in Fig. (2.8).

$$\Omega^{\oplus}$$

$$x = 0 mm$$
Split element at $x = 28.5 mm$

$$x = 200 mm$$

Figure 2.8: FE discretization with Ω^{\ominus} , Ω^{\oplus} and the split element for the numerical simulations. Note that the number of finite element is not respected [Benkemoun et al., 2017a].

2.4.6 Chloride diffusion without chloride binding

For a case of chloride diffusion without chloride binding, a chloride surface concentration c_0 equals to $10^{-2}g/mm^3$ is imposed at x = 0 at the left side of the bar. The diffusion coefficient D^{\oplus} of Ω^{\oplus} and D^{\oplus} of Ω^{\oplus} are such that their ratios (D^{\oplus}/D^{\oplus}) vary as 5,10 and 100.

The finite element results are compared with results obtained from the analytical solution proposed for a two-phase semi-infinite domain (see [Lewis et al., 2004] for instance). The concentrations $c_f^{\ominus}(x,t)$ in Ω^{\ominus} and $c_f^{\oplus}(x,t)$ in Ω^{\oplus} are known such as:

$$c_{f}^{\ominus}(x,t) = c_{0} \sum_{n=0}^{\infty} \tau^{n} \left(\operatorname{erfc} \frac{(2n+1)l^{e} + x}{2\sqrt{(D^{\ominus}t)}} - \tau \operatorname{erfc} \frac{(2n+1)l^{e} - x}{2\sqrt{(D^{\ominus}t)}} \right)$$
(2.63)

and

$$c_f^{\oplus}(x,t) = \frac{2kc_0}{k+1} \sum_{n=0}^{\infty} \tau^n \operatorname{erfc} \frac{(2n+1)l^e + kx}{2\sqrt{(D^{\ominus}t)}}$$
(2.64)

where

$$k = (\frac{D^{\ominus}}{D^{\oplus}})^{0.5}$$
 and $\tau = \frac{1-k}{1+k}$.

Fig. (2.9(a), 2.9(b), 2.9(c)) plot the analytical solution for Ω^{\ominus} and Ω^{\oplus} versus numerical results (E-FEM) for $\frac{D^{\ominus}}{D^{\oplus}}$ equals to 5, 10 and 100, respectively and for t =50, 100, 500, 1000 and 10000 days. For the computation, we choose n = 10. We observe a good agreement between the analytical solution and numerical simulations, notably at $x = 28.5 \ mm$ where the jump in the diffusion coefficient is located. Note that for t = 10000 days, we have not superimposed the analytical solution Eq. (2.63 and 2.64) because for this value of t the hypothesis of a semi-infinite medium is no longer right.

2.4.7 Chloride diffusion with chloride binding

To take the chloride binding capacity in consideration, the values of α_L and β_L are considered as indicated by [Sergi et al., 1992b]. A chloride surface concentration c_0 is imposed at x = 0 and for t > 0 at the left side of the bar. The ratios D^{\oplus}/D^{\oplus} vary as 1,5,10 and 100.

$c_0 (g/mm^3)$	D^{\ominus}/D^{\oplus} (-)	chloride binding	α_L	β_L
10^{-2}	$\{1, 5, 10, 100\}$	yes	1.67	4.08

Table 2.2: Values for the numerical simulations : chloride diffusion with chloride binding.

Fig. (2.10, 2.11(a), 2.11(b) and 2.11(c)) plot the numerical simulation results versus the analytical solution for $\frac{D^{\ominus}}{D^{\oplus}} = 1,5,10$ and 100, respectively and for t = 50,100, 500, 1000 and 10000 days. For the case $\frac{D^{\ominus}}{D^{\oplus}} = 1, D = D^{\ominus} = D^{\oplus}$, the analytical solution is built from Crank's solution Eq. (2.26) with a modified diffusion coefficient



Figure 2.9: Numerical results (E-FEM) versus exact solution for t = 50, 100, 500, 1000 and 10000 days (no chloride binding)

 D_L accounting for the isotherm is:

$$c_f(x,t) = c_0 \left(1 - \operatorname{erf}\left(\frac{x}{2\sqrt{D_L t}}\right) \right), \qquad (2.65)$$

where D_L is

$$D_L = \frac{D}{1 + \frac{\alpha_L}{[1 + \beta_L c(x,t)]^2}}$$
(2.66)

c(x, t) is the typical Crank's solution with a constant diffusion coefficient D and the Langmuir isotherm is chosen here. Again, a good agreement is observed between the numerical simulation results and the analytical solution outputs (2.65). As in previous case, note that for t = 10000 days, we have not superimposed the analytical solution Eq. (2.65).



Figure 2.10: Numerical results versus analytical solution for $\frac{D^{\ominus}}{D^{\oplus}} = 1$ and t = 50, 100, 500, 1000 and 10000 days (chloride binding)

For the ratios $\frac{D^{\ominus}}{D^{\oplus}} = 5$, 10 and 100, the analytical solution is obtained using Eq. (2.63) and (2.64) with a modified coefficient D_L^{\ominus} in Ω^{\ominus} and D_L^{\oplus} in Ω^{\oplus} such as

$$c_{L}^{\ominus}(x,t) = c_{0} \sum_{n=0}^{\infty} \tau^{n} \left(\operatorname{erfc} \frac{(2n+1)l^{e} + x}{2\sqrt{(D_{L}^{\ominus}t)}} - \tau \operatorname{erfc} \frac{(2n+1)l^{e} - x}{2\sqrt{(D_{L}^{\ominus}t)}} \right)$$
(2.67)

and

$$c_L^{\oplus}(x,t) = \frac{2kc_0}{k+1} \sum_{n=0}^{\infty} \tau^n \operatorname{erfc} \frac{(2n+1)l^e + kx}{2\sqrt{(D_L^{\oplus}t)}}$$
(2.68)

where

$$k = \left(\frac{D_L^{\ominus}}{D_L^{\oplus}}\right)^{0.5}$$
 and $\tau = \frac{1-k}{1+k}$

We note

$$D_L^{\ominus(\oplus)} = \frac{D^{\ominus(\oplus)}}{1 + \frac{\alpha_L^{\ominus(\oplus)}}{[1 + \beta_L^{\ominus(\oplus)} c_f^{\ominus(\oplus)}(x,t)]^2}}$$
(2.69)

Fig. (2.11(a), 2.11(b) and 2.11(c)) show a good agreement between the numerical simulation results (E-FEM) and the analytical solutions Eq. (2.67) and (2.68)).

Furthermore, in order to assess the role of chloride binding integration with chloride diffusion properties, a comparison between the concentration profiles with chloride binding ("cb") and without chloride binding ("ncb") has been done. Fig. (2.12(a), 2.12(b) and 2.12(c)) show the concentration profiles for $\frac{D^{\ominus}}{D^{\oplus}} = 5$, 10 and 100 and t = 100, 500, 1000 and 10000 days with and without chloride binding. We note that the amount of free-chlorides (chlorides in solution) that are responsible for reinforcement corrosion is less important when chloride binding is considered. This conclusion is similarly figured out in [Martin-Perez et al., 2000] throughout a one dimensional finite difference model where both no isotherm and linear-non linear isotherms relations are considered. Consequently for durability issues, it is relevant to consider chloride binding for not overestimating the time span until corrosion initiation and consequently for accurately estimating the service life of a structure.



Figure 2.11: Numerical results (E-FEM) versus exact solution for t = 50, 100, 500, 1000 and 10000 days (chloride binding)



Figure 2.12: Influence of the chloride binding on the chloride concentration for t = 100, 500, 1000 and 10000 days

2.4.8 Evaluation of the corrosion initiation time t_{\circ}

To examine the effect of chloride binding isotherm, we will plot the evolution of c_f in relation with time at x equals to 40 mm, corresponding to the concrete cover d_c . The initiation of corrosion begins when the concentration of chloride around the reinforcement exceeds the threshold chloride concentration (C_{th}) . When chloride binding is not considered in calculation, Fig. (2.13) plots the evolution of c_f in relation with time t for different ratios $\frac{D^{\ominus}}{D^{\oplus}}$. The parameters for the study are chosen from [Liang et al., 2009] and are summarized in Table 2.3. The function f(t) for the numerical results fitting is chosen such as [Benkemoun et al., 2017a]:

$$f(t) = A \operatorname{erfc}(\frac{B}{t^n}).$$
(2.70)

where A, B and n are constant values depending on $\frac{D^{\ominus}}{D^{\oplus}}$ and erfc the complementary error function. Fig. (2.13) shows that the corrosion initiation period t_{\circ} equals to 210 days, 142 days and 76 days for $\frac{D^{\ominus}}{D^{\oplus}} = 5$, 10 and 100, respectively.

c_{th}/c_0 (-)	$d_c \ (mm)$	α_L	β_L
0.32	40	1.67	4.08

Table 2.3: Parameters for the computation of the corrosion initiation time (from [Liang et al., 2009])

However, the evolution of corrosion initiation time t_{\circ} in function of $\frac{D^{\ominus}}{D^{\oplus}}$ and the chloride binding capacity is shown in Fig. (2.14(a), 2.14(b) and 2.14(c)). They show the evolution of c_f , with ("cb") or without chloride binding ("ncb"), in relation with time t. The parameters used in this case are chosen from [Liang et al., 2009] and summarized in Table 2.3.

Fig. (2.14(a), 2.14(b) and 2.14(c)) show that the corrosion initiation period t_{\circ}



Figure 2.13: Evolution of the concentration c_f in relation with time t for different ratios $\frac{D^{\ominus}}{D^{\oplus}}$ with the corresponding concentration threshold for t_{\circ}

equals to 556 days, 367 days and 198 days for $\frac{D^{\ominus}}{D^{\oplus}} = 5$, 10 and 100, respectively. The results are sum up in Table 2.4. We can observe that considering the chloride binding capacity for the study of service life of concrete structures is relevant. It removes chloride from the diffusion flux, thus retarding the time span until corrosion initiation as it is illustrated in the numerical results.

D^{\ominus}/D^{\oplus} (-)	$t_{\circ} \text{ ncb (days)}$	$t_{\circ} \text{ cb (days)}$
5	210	556
10	142	367
100	76	198

Table 2.4: Values for t_{\circ} in relation with $\frac{D^{\ominus}}{D^{\oplus}}$ and the chloride binding capacity

2.5 Conclusion

In this chapter, a numerical FE solution of the mass transport problem considering diffusivity (using Fick's second law), migration (using Nernst-Planck equation) and chloride binding (using nonlinear isotherm) is firstly presented by a classical finite element method.



Figure 2.14: Evolution of the concentration c_f in relation with time t with the corresponding concentration threshold t_{\circ}

However, the meso-structure of concrete is considered as a mixture of heterogeneous materials with aggregates embedded in a mortar matrix .In order to take into account these heterogeneities without any mesh adaptation. A weak discontinuity is introduced into the strain field. The mathematical and numerical framework of the E-FEM was presented as an effective method to account for the kinematic enhancement.

Calibration of the finite element model with analytic solutions have been carried out considering different transport mechanisms. The comparison was implemented also for the two-phase element modeling of a simple diffusion problem accounting for chloride binding. A good agreement between analytical solution and numerical simulations has been observed in this comparison.

The evaluation of the time span until corrosion initiation is evaluated Knowing the concentration profiles evolution for different time interval values. Then, it is noted that t_{\circ} is :

- retarded when chloride binding capacity effects are considered;
- accelerated when $D_{matrix}/D_{aggregate}$ is increased.

Consequently, taking explicitly into account the microstructure diffusion coefficients and the chloride binding effects are important to consider for a better prediction of the time span until corrosion initiation.

In the next chapter, we turn to 3D simulations. A homogenization method combined to the E-FEM will be used in order to:

• determine Representative Volume Element through numerical homogenization;

• compute macroscopic diffusivity tensors accounting for the microstructure diffusivity properties.

Chapter 3

3D homogenization of chloride transport in uncracked concrete

3.1 Introduction

It has currently been noticed that the strength-based design cannot ascertain a reliable estimation of the service life of concrete structures. This means that durability is a critical aspect in the rational design of concrete structures. The chloride ion diffusion coefficient is the transport property that is considered as the most important information on durability design and service life prediction [Wu et al., 2016]. Note that in the design of dams and other large hydraulic structures, permeability is also one of the most important transport properties affecting the durability and serviceability of concrete structures [Li et al., 2016].

However, concrete has a complex mesostructure that contains inclusions embedded in the mortar (or cement paste) as well as the ITZ between them. Transport properties are concequently thoroughly affected by the interrelating parameters associated with the mesoscopic composition which is difficult to assess through experimental procedures [Abyaneh et al., 2013]. Consequently, this complexity makes it difficult to determine the realistic diffusivity of concrete. For this reason, it would be essential to estimate the transport properties of concrete depending on its mesostructure, using analytical or numerical models. While, classic analytical models presented in literature generally involve predicting of transport properties according to concrete mixture and cannot account for complex geometrical arrangements and consequently unable to explain the complex transport behavior of concrete. Some of these models like the Parallel model, the Series model, the Maxwell-Eucken (ME) model and the Effective Medium Theory (EMT) model are summarized in [Wang and Pan, 2008].

The reliability and efficiency of the recent numerical analysis tools made it suitable to simulate the composite behavior of concrete at mesoscopic level. Modeling chloride ions transport with a mesoscale model in the context of the previously presented Embedded Finite Element Methods can be regarded as a successful and alternative numerical approach for considering the microstructure in the determination of transport properties.

In this chapter, the attention will be paid to the implementation of a homogenization method which is described later in this chapter and integrated to the Embedded Finite Element Method.

The finite element formulation is built through a discrete (lattice) finite element
model and the meso-structure is based on a two-phase 3D representation of heterogeneous materials, such as concrete, where stiff aggregates are embedded into a mortar matrix.

Herein, the suitable information of chloride ions transport and interactions between the different phases of the material at the meso-scale are used to homogenize the behavior and to compute effective concrete transport properties.

The remainder of this chapter is organized as follows. The homogenization method retained to compute effective diffusivity tensor at the macroscale is described in Section 2. The suggested method of computation leads to an effective diffusivity coefficient accounting for chloride binding even in steady-state conditions is detailed in Section 3. In Section 4, computational homogenization is used to determine effective diffusivity tensor in RVE containing microstructure embedded through the E-FEM. Finally, the comparison with the result of Maxwell's equation is carried out regarding the case of no binding effect. Comparison with experimental work is also performed to show the applicability of the proposed numerical approach regarding the case of binding effect.

3.2 Governing equation and Embedded Finite Element formulation

The homogenization method mentioned above is used to perform the numerical simulations the calculation of macroscopic diffusivity tensors. To realize this calculation, a steady-state condition is supposed to achieve a steady flux with no variation of chloride ions with time. Steady-state condition is considered here as a relevant issue following the long term chloride exposure considerations in durability problems.

However, if the chloride binding is considered in Fick's Law, one gets:

$$\frac{\partial c_t}{\partial t} = \frac{\partial c_f}{\partial t} + \frac{\partial c_b}{\partial t} = D_e \frac{\partial^2 c}{\partial x^2}$$
(3.1)

Therefore, Fick's second law of diffusion can not be used to obtain steady flux taking into account the effect of chloride binding in the simulations. In order to overcome this drawback, we used the study of Carrara et al [Carrara et al., 2016], that leads to an effective diffusivity coefficient accounting for chloride binding even in steady-state conditions.

3.2.1 Governing equation

To make it possible to to take into account chloride binding capacity when steadystate conditions are assumed, we consider the total chloride ions concentration c_t instead of considering free chloride ions concentration c_f as the problem variable.

Starting from

$$\frac{\partial c_t}{\partial t} - \nabla \cdot (D\nabla(c_f)) = 0, \qquad (3.2)$$

we can now consider the relationship

$$\nabla(c_f) = \nabla(c_t - c_b) = (1 - \frac{\partial c_b}{\partial c_t})\nabla(c_t).$$
(3.3)

Equation (3.2) with Eq. (3.3) becomes:

$$\frac{\partial c_t}{\partial t} - \nabla \cdot \left(\left(1 - \frac{\partial c_b}{\partial c_t}\right) D \nabla(c_t) \right) = 0.$$
(3.4)

As in [Carrara et al., 2016], we define $\mu_B(c_t)$ such as

$$\mu_B(c_t) = \left(1 - \frac{\partial c_b}{\partial c_t}\right) \tag{3.5}$$

leading to the definition of an effective diffusivity coefficient $D_{eff}(c_t)$ such as

$$D_{eff}(c_t) = \left(1 - \frac{\partial c_b}{\partial c_t}\right)D.$$
(3.6)

Consequently Eq. (3.4) becomes

$$\frac{\partial c_t}{\partial t} - \nabla \cdot (D_{eff}(c_t)\nabla(c_t)) = 0.$$
(3.7)

However, the amount of chloride ions bound c_b expressed, in general, by isotherm models as a function of c_f instead of c_t , i.e $c_b = c_b(c_f)$. Hence, a little modification has to be done for $\mu_B(c_t)$ so that it can express the derivative of c_b in function of c_f :

$$\mu_B = (1 - \frac{\partial c_b}{\partial c_t}) = \frac{\partial c_f}{\partial c_t} = \left[\frac{\partial (c_b + c_f)}{\partial c_f}\right]^{-1} = (1 + \frac{\partial c_b}{\partial c_f})^{-1}$$
(3.8)

where now μ_B is a function of c_f and the classical isotherm models can be used.

Finally Eq. (3.7) becomes

$$\frac{\partial c_t}{\partial t} - \nabla \cdot \left(D_{eff}(c_f) \nabla(c_t) \right) = 0, \qquad (3.9)$$

In conclusion, even if steady-state conditions are now considered the chloride binding capacity is still accounted for in the governing equation through the effective diffusivity tensor $D_{eff}(c_f)$ and the problem to be solved is:

$$\nabla \cdot (D_{eff}(c_f)\nabla(c_t)) = 0. \tag{3.10}$$

The consideration of chloride binding in steady state means that all the sites in cementitious matrix which can bound the chloride are not saturated. This state is called "pseudo-steady" [Amiri et al., 1997].

3.2.1.1 Galerkin approximation

The Galerkin approximation of Eq. (3.9) leads to

$$\int_{\Omega} \delta c_t \frac{\partial c_t}{\partial t} d\Omega + \int_{\Omega} \frac{\partial \delta c_t}{\partial x} D_{eff}(c_f) \frac{\partial c_t}{\partial x} d\Omega - \int_{\partial \Omega_{J_{c_t}}} \delta c_t \bar{J}_{c_t} d\partial \Omega = 0, \quad (3.11)$$

where δc_t is the virtual total chloride concentration in the space \mathcal{C} such as $\mathcal{C} = \{\delta c_t(x) : \Omega \mapsto \mathbb{R} \mid \delta c_t \in H^1, \delta c_t = 0 \text{ on } \partial \Omega_{c_t}\}$. We note $\partial \Omega_{c_t}$ the part of the boundary where the total chloride concentration is imposed and $\partial \Omega_{J_{c_t}}$ where the flux \overline{J}_{c_t} is imposed.

3.2.1.2 Discretization of the total chloride concentration field

The discretization of c_t and δc_t is made as in chapter 2 and referring to [Benkemoun et al., 2015] leading to:

$$c_t = \mathbf{N}\bar{\mathbf{c}}_t^h + M^{\oplus/\ominus}\bar{\bar{c}}_t^h, \qquad (3.12)$$

$$\delta c_t = \mathbf{N} \delta \bar{\mathbf{c}}_t^h + M^{\oplus/\Theta} \delta \bar{\bar{c}}_t^h, \qquad (3.13)$$

and

:

$$\frac{\partial c_t}{\partial x} = \boldsymbol{B}\bar{\boldsymbol{c}}_t^h + G^{\oplus/\ominus}\bar{\bar{c}}_t^h, \qquad (3.14)$$

$$\frac{\partial \delta c_t}{\partial x} = \boldsymbol{B} \delta \bar{\boldsymbol{c}}_t^h + G^{\oplus/\Theta} \delta \bar{\bar{c}}_t^h.$$
(3.15)

Considering Eq. (3.12), (3.13), (3.14), (3.15) and (3.11) for any $\delta \bar{c}_t^h$ and $\delta \bar{c}_t^h$ yield

$$\begin{cases} \mathcal{M}\dot{\bar{c}}_{t}^{h} + \mathcal{P}\dot{\bar{c}}_{t}^{h} + \mathcal{K}(c_{f})\bar{c}_{t}^{h} + \mathcal{F}(c_{f})\bar{\bar{c}}_{t}^{h} = \boldsymbol{f}^{ext} \\ \mathcal{P}^{T}\dot{\bar{c}}_{t}^{h} + \mathcal{L}\dot{\bar{c}}_{t}^{h} + \mathcal{F}^{T}(c_{f})\bar{c}_{t}^{h} + \mathcal{H}(c_{f})\bar{\bar{c}}_{t}^{h} = 0 \end{cases}$$
(3.16)

where (\bullet) represents time derivatives and where the flux \bar{J}_{c_f} is applied only on the regular part of the total chloride concentration field. We note

$$\mathcal{M} = \int_{\Omega} \mathbf{N}^{T} \mathbf{N} d\Omega, \qquad \mathcal{P} = \int_{\Omega} \mathbf{N}^{T} M^{\oplus/\Theta} d\Omega \qquad (3.17)$$

$$\mathcal{K}(c_f) = \int_{\Omega} \boldsymbol{B}^{\boldsymbol{T}} D_{eff}(c_f) \boldsymbol{B} d\Omega, \qquad \mathcal{F}(c_f) = \int_{\Omega} \boldsymbol{B}^{\boldsymbol{T}} D_{eff}(c_f) G^{\oplus/\Theta} d\Omega \qquad (3.18)$$

$$\mathcal{L} = \int_{\Omega} M^{\oplus/\ominus} M^{\oplus/\ominus} d\Omega, \qquad \mathcal{H}(c_f) = \int_{\Omega} G^{\oplus/\ominus} D_{eff}(c_f) G^{\oplus/\ominus} d\Omega$$
(3.19)

$$\boldsymbol{f}^{ext} = \int_{\partial\Omega_{J_{c_t}}} \boldsymbol{N}^T \bar{J}_{c_t} d\partial\Omega, \qquad (3.20)$$

and $D_{eff}(c_f)$ such as

$$D_{eff}(c_f) = \frac{D^{\oplus/\ominus}}{\left(1 + \left(\frac{\partial c_b}{\partial c_f}\right)^{\oplus/\ominus}\right)}$$
(3.21)

The Langmuir isotherm model is considered in this step to represent the relation $\left(\frac{\partial c_b}{\partial c_f}\right)$ such as:

$$\left(\frac{\partial c_b}{\partial c_f}\right)^{\oplus/\ominus} = \frac{\alpha_L^{\oplus/\ominus}}{(1 + \beta_L^{\oplus/\ominus} c_f^{\oplus/\ominus})^2} \tag{3.22}$$

Finally Eq. (3.16) have to be coupled to

$$c_f^{\oplus/\ominus} + c_b^{\oplus/\ominus}(c_f) = c_t^{\oplus/\ominus}$$
(3.23)

in order to compute $c_f^{\oplus/\ominus}$, then $D_{eff}(c_f)$ and consequently to update Eq. (3.16).

To conclude, on the basis of the work of [Carrara et al., 2016], we have suggested a FE formulation based upon the E-FEM similar to that shown in chapter 2, in order to take into account the chloride binding capacity in a two-phase material. The key point being that both in steady-state and transient conditions the chloride binding capacity is taken into account.

3.3 Method of homogenization

Upscaling, or homogenization, can be identified as the procedure of substituting some heterogeneous properties at fine scale like: meso, micro or nano scale, with an equivalent homogeneous property at macro scale. For example, the assessed macroscopic transport properties of concrete are supposed to be dependent on the transport properties and content of aggregate in addition to the composition and porosity of cement paste. So, it stands to reason that effective property value of coarse-scale model varies according to the variation of sub-scale features.

Many homogenization techniques were derived to provide such properties. Some authors predicted the homogenized chloride diffusivity of concrete as two or threephases material by taking into account the morphological characteristics and the physical properties (like transport properties and volume fraction) of each phase constituent material using analytical methods [Hobbs, 1999, Zheng and Zhou, 2008, Savija et al., 2013], (see chapter one). This approach considers just one characteristic of the respective constituent and the volume ratio of the heterogeneities, but neglects the effect of other aspects.

Other works developed multiscale computational homogenization models where they were used with success in mechanics and civil engineering domains. In general, we can distinguish two main types of homogenization methods:

- The averaging technique of homogenization [Samson et al., 1999, Samson et al., 2005].
- Periodic homogenization technique [Mchirgui et al., 2016, Bourbatache et al.,

2012]

In this work, the spatial averages is used to compute macroscopic diffusivity tensor by applying several calculations solved on a mesoscale level under well defined boundary conditions [Pouya and Fouché, 2009, Jourdain et al., 2014a].

3.3.1 Mean gradient of concentration and mean flux

In this part, we only focus on transport by diffusion in fully saturated conditions. The chloride ions transport model is governed by Fick's Law :

$$\vec{q}(\vec{x}) = -D_m \cdot \vec{\nabla} c(\vec{x}) \tag{3.24}$$

where $\vec{q}(\vec{x})$ is the mass flux $[kg/(m^2s)]$, D_m the mesoscale diffusion coefficient $[m^2/s]$, $\vec{\nabla}$ the gradient operator [1/m] and $c(\vec{x})$ the mass concentration $[kg/m^3]$. Inserting Eq. (3.24) in the mass balance equation [Poulsen and Mejlbro, 2010], Fick's second Law is obtained and considered as the problem to be solved in terms of mass concentration field.

The macroscopic diffusivity for the domain Ω is defined by a positive-definite symmetric tensor $\underline{\mathbf{D}}$ which represents a direct link between the mean concentration gradient \vec{G} and the mean flux \vec{Q} such as:

$$\vec{Q} = -\underline{\mathbf{D}} \cdot \vec{G} \tag{3.25}$$

Herein, the mean concentration gradient $\vec{G} [kg/m^4]$ and the mean flux $\vec{Q} [kg/(m^2s)]$ within a domain Ω is defined as spatial averages of the scalars corresponding to the mesoscale by the following relationships:

$$\vec{G} = \frac{1}{V} \int_{\Omega} \vec{\nabla} c(\vec{x}) d\Omega, \qquad (3.26)$$

$$\vec{Q} = \frac{1}{V} \int_{\Omega} \vec{q}(\vec{x}) d\Omega, \qquad (3.27)$$

where V is the volume of Ω . We remind that \vec{q} is Fick's velocity at point \vec{x} and $\vec{\nabla}c(\vec{x})$ is the concentration gradient at this point such as the law of behavior on the mesoscale (Fick's Law) gives:

$$\vec{q}(\vec{x}) = -D_m \cdot \vec{\nabla} c(\vec{x}) \tag{3.28}$$

where D_m is the mesoscale diffusion coefficient. In order to obtain a matrix which can represent anisotropic diffusivity, the law of behavior on the fine scale must be a function of the opening and the orientation of the cracks. The law we use relates the coefficient of diffusion to the crack width obtained from the mechanical model calculations. This consideration relies on the experimental results of [Djerbi et al., 2008] which was presented in chapter one. Djerbi et al. provide the relation between the diffusion coefficient through the crack D_{cr} and the crack width values [|u|] according to their experimental results such as:

$$\begin{cases} D_{cr}(m^2/s) = 1.8 \times 10^{-12}, & [|u|] < 30 \,\mu m \\ D_{cr}(m^2/s) = 2 \times 10^{-11} [|u|] - 4 \times 10^{-10}, & 30 \,\mu m \le [|u|] \le 80 \,\mu m \\ D_{cr}(m^2/s) = 14 \times 10^{-10}, & [|u|] > 80 \,\mu m \end{cases}$$
(3.29)

where D_{cr} is nothing but the coefficient of diffusion at mesoscale D_m for cracked

element.

We have to note that the cracking effect on the diffusion of chloride will be studied in the next chapter.

Pouya et al. [Pouya and Courtois, 2002, Pouya and Fouché, 2009] proposed a method to compute the macroscopic permeability tensor for hydraulic transport problem (incompressible flow) in heterogeneous media. This method was developed by Jourdan et al. [Jourdain et al., 2014a] for gas transport problem (compressible flow) in cracked media such as concrete. The same method is adopted here to calculate the macroscopic diffusivity tensor in heterogeneous cracked media. The volumetric mean values of the flux \vec{Q} and the concentration gradient \vec{G} can be computed from the boundary values of concentration and flux on $\partial\Omega$. Green's theorem is used to obtain:

$$\vec{G} = \frac{1}{V} \int_{\partial \Omega} c(\vec{x}) \, \vec{n}(\vec{x}) \, dS, \qquad (3.30)$$

$$\vec{Q} = \frac{1}{V} \int_{\partial \Omega} (\vec{q} \cdot \vec{n}) \, \vec{x} \, dS, \tag{3.31}$$

 \vec{n} represents the outward unit vector from the surface S and dS the surface element on $\partial\Omega$. The last equations [3.30,3.31] define the spatial averages of the chloride mesoscale flow and concentration on $d\Omega$. They can provide a sequence of numerical simulation to compute the macroscopic diffusivity tensor in the presence of cracks and inclusions for any domain shape and any boundary conditions [Pouya and Courtois, 2002].

3.3.2 Boundary conditions: linear concentration

In the upscaling methods developed for such transport problems, two types of boundary conditions are considered: (1) a permeameter boundary conditions where the flux is imposed in one main direction and no fluxes are imposed in other directions [Bailly et al., 2009], (2) a linear pressure boundary conditions at the contour. The linear boundary conditions is represented in Fig. (3.1) assuming a rectangular shape in the two-dimensional case [Long et al., 1982]. By following the method proposed in [Pouya and Courtois, 2002], a linear concentration condition is applied on the contour as a Dirichlet-type (see Fig. (3.1)) boundary conditions such as:

$$c(\vec{x}) = \vec{A} \cdot \vec{x} + c_0, \quad \forall \vec{x} \in \partial \Omega$$
(3.32)

where \vec{A} is a constant vector and c_0 a constant scalar.

The development proposed in Pouya et al. [Pouya and Courtois, 2002] for hydraulic transport problem under the above-mentioned condition leads to the equality $\vec{G} = \vec{A}$. Consequently, equation 3.25 can be written as:

$$\vec{Q} = -\underline{\mathbf{D}} \cdot \vec{A} \tag{3.33}$$

This computational method provides a straightforward way for the numerical computation of the macroscopic diffusivity tensor $\underline{\mathbf{D}}$. To compute the nine components of the tensor $\underline{\mathbf{D}}$, the mean flux gradient \vec{Q} is computed for three distinct directions of the vector \vec{A} . These directions are shown in Fig. (3.2) for a



Figure 3.1: linear concentration boundary condition for a square domain [Pouya and Fouché, 2009].

 $100 \times 100 \times 100 \quad mm^3$ domain: Fig. (3.2(a)) corresponds to X direction, Fig. (3.2(b)) to the Y direction and Fig. (3.2(c)) to the Z direction of the domain.



Figure 3.2: Condition of linear concentration on the contour in the X, Y and Z directions [Benkemoun et al., 2017b].

3.4 3D meso-scale simulations and analysis

3D mesostructures with random aggregates are set as numerical example of concrete in this section. The aggregate particles range from 4 to 20 mm in computational with 20, 30 and 44 % of volumic fraction are embedded in a $100 \times 100 \times 100 \text{ mm}^3$ computational cube as shown in Fig. 3.3. We note D_m the diffusivity in the mortar matrix and D_a the diffusivity in the aggregate particles.



(c) targeted volume fraction $f\approx 45~\%$

Figure 3.3: 3D mesostructure of concrete generated by randomly placing aggregate particles ranging from 4 to 20 mm in a $100 \times 100 \times 100 \text{ mm}^3$ computational cube. The element split into two parts are depicted in dark color and the elements belonging to the aggregates in light grey. Note that the cement paste is not represented for sake of clarity [Benkemoun et al., 2017a].

3.4.1 Mesostructure and RVE determination

The aforementioned numerical homogenization method has been used for the numerical determination of a Representative Volume Element(RVE) to estimate the homogenized properties of heterogeneous materials like effective diffusivity tensors \underline{D} [Benkemoun et al., 2017a].

In this work, the RVE classical definition proposed in [Kanit et al., 2003] is introduced. The RVE can be defined that for a given cement-based material, it is large enough and contains a sufficient number of heterogeneities for the macroscopic moduli to be independent of the boundary conditions [Sab, 1992]. In order to numerically determine RVE, two convergence criteria proposed in [Li et al., 2016] are regarded :

- The estimated properties of RVE are independent from realizations in which aggregates particles are randomly placed. In other words, the CoV of diffusivity coefficients from these realizations achieves a given accuracy, ϵ_1
- The RVE is large enough to be statistically representative of the overall concrete diffusivity such as :

$$\epsilon_{L_s} = \left| \frac{D(L_s) - D_t}{D_t} \right| \le \epsilon_2 \tag{3.34}$$

where $D(L_s)$ is the diffusivity coefficient of the 3D concrete sample sized by L_s , D_t is the overall diffusivity for the concrete and ϵ_2 the given error.

Based on these criteria, numerical simulations on 3D samples sized by L_s are carried out to determine RVE. Concerning the values of L_s for the simulations, we refer to the work of [Abyaneh et al., 2013]. Indeed, it is stated in [Abyaneh et al., 2013] that a computational cube that is 2.5 times the size of the largest aggregate can give representative results if a sufficient number of realizations are considered and then the results averaged. In this sense, three sizes L_s for the computational cube are regarded : 50 mm, 80 mm and 100 mm. Note that we assume a constant number of nodes - 630000 - for the computational cubes leading to a typical element size equals to 0.5 mm, 0.8 mm and 1.0 mm for $50 \times 50 \times 50 \text{ mm}^3$, $80 \times 80 \times 80 \text{ mm}^3$ and $100 \times 100 \times 100 \text{ mm}^3$, respectively.

3.4.1.1 Numerical results and discussion

Table 3.2, 3.3 and 3.4 show the convergence criteria values computed from the numerical simulations for L_s equals to 50 mm, 80 mm and 100 mm, respectively. Table 3.1 sums up the values retained for the simulations and for the convergence criteria.

number of realizations	targeted volumic fraction $(\%)$	ϵ_1 (from [Li et al., 2016])	ϵ_2 (from [Li et al., 2016])
100	35.5	3.00 E-03	4.00 E-03

Table 3.1: Numerical values for the simulations and the convergence criteria

Diagonal components	numerical results	
Diagonal components	CoV	ϵ_{L_s}
D_{xx}/D_m	3.52 E-03	6.58 E-03
D_{yy}/D_m	3.32 E-03	8.89 E-03
D_{zz}/D_m	3.80 E-03	8.70 E-03

Table 3.2: Convergence criteria values for $50 \times 50 \times 50 mm^3$ with 35 % of volumic fraction. Typical element size equals to 0.5 mm

A decreasing in the CoV of diffusion coefficient is observed when increasing the sample size. It can be stressed that for L_s equals to 50 mm - limit size of representativeness regarding the largest size of aggregates - the CoV of diffusivity coefficients is close to (almost below) the given error ϵ_1 . Regarding the relative error

Diagonal components	numerical results	
Diagonai componentis	CoV	ϵ_{L_s}
D_{xx}/D_m	1.93 E-03	9.98 E-03
D_{yy}/D_m	2.24 E-03	1.33 E-02
D_{zz}/D_m	2.04 E-03	1.41 E-02

Table 3.3: Convergence criteria values for $80 \times 80 \times 80 \ mm^3$ with 35 % of volumic fraction. Typical element size equals to 0.8 mm

Diagonal components	numerical results	
Diagonai componentis	CoV	ϵ_{L_s}
D_{xx}/D_m	1.32 E-03	1.15 E-02
D_{yy}/D_m	1.50 E-03	1.52 E-02
D_{zz}/D_m	1.16 E-03	1.50 E-02

Table 3.4: Convergence criteria values for $100 \times 100 \times 100 \ mm^3$ with 35 % of volumic fraction. Typical element size equals to 1.0 mm

 ϵ_{L_s} , we observe that (1) for L_s equals to 50 mm, ϵ_{L_s} is not inferior to the given error ϵ_2 and (2) as the sample size increases, the values of ϵ_{L_s} increase. These two issues can be justified by the typical finite element size which is increased from 0.5 to 1 mm as the sample size increases. Consequently, the typical finite element size has to be reduced below 0.5 mm to achieve a value below the given error ϵ_2 . Table 3.5 presents the convergence criteria values computed from the numerical simulations for L_s equals to 50 mm with a typical element size now equals to 0.35 mm. Note that we cannot unfortunately consider an element size below 0.35 mm to keep a reasonable computational time. The relative error is now below the given error ϵ_2 . Consequently, the 50×50×50 mm³ domain with a typical element size equals to 0.35 mm to 0.35 mm is considered as a RVE for the determination of diffusivity tensors.

Diagonal components	numerical results	
Diagonal components	CoV	ϵ_{L_s}
D_{xx}/D_m	2.72 E-03	1.90 E-03
D_{yy}/D_m	2.95 E-03	7.12 E-04
D_{zz}/D_m	2.82 E-03	1.18 E-03

Table 3.5: Convergence criteria values for $50 \times 50 \times 50 \ mm^3$ with 35 % of volumic fraction. Typical element size equals to 0.35 mm

3.4.2 Macroscopic diffusivity tensor computation

Herein, we show numerical results to examine the implementation of the homogenization method proposed in Section 3.3. The numerical results include both diffusivity tensors with and without chloride binding effect. The symmetric and positive-definiteness of the computed diffusivity tensors are proved hereafter. The E-FEM model incorporated in the meso-macro approach is validated in both simulation and experimental results. The comparison with the result of Maxwell's equation is carried out regarding the case of no binding effect. However, a comparison with results obtained by experiments is done to check the numerical approach validity. A sensitivity analysis is implemented to examine the effect of chloride binding isotherm parameters on the computed diffusivity tensors.

3.4.3 Diffusivity tensor results

The numerical results of macroscopic diffusivity tensor is presented hereafter considering D_a/D_m ratio as: 0.5, 1.0 and 2.0, where D_a represents the aggregate diffusion coefficient and D_m the mortar diffusion coefficient. The dimension of the numerical sample is chosen according to the previously mentioned RVE as $(50 \times 50 \times 50 \text{ mm}^3)$. The volume fraction of inclusions equals to 35%. The components of diffusivity tensor without accounting for chloride binding effect is presented such as:

$$\underline{\underline{\underline{D}}}_{\overline{D}_{m}} = \begin{pmatrix} 0.793 & -3.159 \times 10^{-4} & 5.351 \times 10^{-4} \\ -3.160 \times 10^{-4} & 0.795 & -5.256 \times 10^{-4} \\ 5.351 \times 10^{-4} & -5.256 \times 10^{-4} & 0.795 \end{pmatrix}_{D_{a}/D_{m}=0.5}$$

$$\underline{\underline{D}}_{\overline{D}_{m}} = \begin{pmatrix} 0.992 & 1.566 \times 10^{-6} & 3.758 \times 10^{-6} \\ 1.558 \times 10^{-6} & 0.994 & 6.124 \times 10^{-8} \\ 3.772 \times 10^{-6} & 4.494 \times 10^{-8} & 0.992 \end{pmatrix}_{D_{a}/D_{m}=1.0}$$

$$\underline{\underline{D}}_{\overline{D}_{m}} = \begin{pmatrix} 1.290 & -6.427 \times 10^{-4} & 1.146 \times 10^{-3} \\ -6.427 \times 10^{-4} & 1.291 & -1.174 \times 10^{-3} \\ 1.146 \times 10^{-3} & -1.174 \times 10^{-3} & 1.294 \end{pmatrix}_{D_{a}/D_{m}=2.0}$$

To affirm the positive-definiteness of the tensor, we compute the eigenvalues. The tensors eigenvalues set is positive such as: $(0.793, 0.796, 0.795)_{D_a/D_m=0.5},$ $(0.992, 0.992, 0.994)_{D_a/D_m=1.0}$ and $(1.290, 1.291, 1.295)_{D_a/D_m=2.0}.$

In order to prove that the tensors are symmetric, we compute :

$$r_F = \frac{||\underline{\underline{A}}_{\underline{\underline{D}}}||_F}{||\underline{\underline{S}}_{\underline{\underline{D}}}||_F}$$
(3.35)

where $|| \bullet ||_F$ is the Frobenius norm, $\underline{\underline{A}}_{\underline{\underline{D}}}$ the anti-symmetric part of $\underline{\underline{D}}$ and $\underline{\underline{S}}_{\underline{\underline{D}}}$ the symmetric part of $\underline{\underline{D}}$ [Benkemoun et al., 2017a]. The Frobenius norm can be defined such as:

$$||\underline{\underline{Q}}||_F = \sqrt{\sum_{i=1}^n \sum_{j=1}^n |Q_{ij}|^2}$$
(3.36)

where $Q = \{Q_{ij}\}$ is a $n \times n$ matrix with real coefficients.

 $\underline{\underline{S}}_{\underline{\underline{Q}}}$ is the symmetric part of $\underline{\underline{Q}}$ such as:

$$\underline{\underline{S}}_{\underline{\underline{Q}}} = \frac{1}{2} (\underline{\underline{Q}} + \underline{\underline{Q}}^T)$$
(3.37)

And $\underline{\underline{A}}_{\underline{Q}}$ the anti-symmetric part of $\underline{\underline{Q}}$ such as:

$$\underline{\underline{A}}_{\underline{\underline{Q}}} = \frac{1}{2} (\underline{\underline{Q}} - \underline{\underline{Q}}^T)$$
(3.38)

Table 3.6 shows the values of r_F as a function of D_a/D_m for both studied cases. In this way, the prevailing role of $\underline{\underline{D}}$'s symmetric part can be figured out for the case of no binding and less for case of binding contribution.

		D_a/D_m	
	0.5	1.0	2.0
r_F (no binding)	5.14 E-08	9.43 E-09	0.00

Table 3.6: Values of r_F as a function of D_a/D_m

In addition, considering the fact that aggregate particles are placed randomly in the computational cube, the tested mesostructures are expected to exhibit identical diffusivity values along the three principal axes and can be represented as a single scalar value, denoted as D_s . For a volume fraction of aggregates corresponding to 35.5 %, we choose D_s/D_m with a value of 0.795, 0.993 and 1.292 for D_a/D_m equals to 0.5, 1.0 and 2.0.

Furthermore, when the contribution of the effect of binding are considered, the complete form of the three diffusivity tensors are:

$$\underline{\underline{\underline{D}}}_{\overline{D_m}} = \begin{pmatrix} 0.603 & 2.367 \times 10^{-4} & 4.279 \times 10^{-4} \\ 4.609 \times 10^{-4} & 0.602 & -5.582 \times 10^{-5} \\ 1.459 \times 10^{-4} & -1.142 \times 10^{-4} & 0.603 \end{pmatrix}_{D_a/D_m=0.5}$$

$$\underline{\underline{\underline{D}}}_{\overline{D_m}} = \begin{pmatrix} 0.782 & -7.817 \times 10^{-4} & 1.068 \times 10^{-4} \\ -1.657 \times 10^{-6} & 0.779 & -3.817 \times 10^{-4} \\ 6.34 \times 10^{-4} & -4.244 \times 10^{-4} & 0.782 \end{pmatrix}_{D_a/D_m = 1.0}$$

$$\underline{\underline{\underline{D}}}_{\overline{D_m}} = \begin{pmatrix} 1.006 & -3.169 \times 10^{-3} & 1.08 \times 10^{-3} \\ -6.761 \times 10^{-4} & 0.999 & -2.015 \times 10^{-3} \\ 3.082 \times 10^{-3} & -2.744 \times 10^{-3} & 1.011 \end{pmatrix}_{D_a/D_m=2.0}$$

The value of D_s obtained from the three diffusivity tensors which are computed for D_a/D_m equals to 0.5, 1.0 and 2.0 such as 0.603, 0.781 and 1.01 respectively.

For this case, the three tensors eigenvalues set is obtained $(0.604, 0.603, 0.601)_{D_a/D_m=0.5},$ $(0.783, 0.782, 0.779)_{D_a/D_m=1.0}$ as: and $(1.013, 1.006, 0.999)_{D_a/D_m=2.0}$. They are also positive and then the matrix is proved to be positive-definite for both cases -with and without binding effect-. However, symmetry of the tensors is not assured when the isotherm parameter β is increased inducing a strong non-linearity. Table 3.7 shows the values of r_F to examine the symmetry of diffusion coefficient tensors corresponding to different values of β . The increase of β would induce a strong non-linearities.

		$\alpha = 1.67$		
β	0.0	0.1	1.0	4.08
r_F	1.09 E-08	4.11 E-06	1.81 E-05	2.34 E-04

Table 3.7: Values of r_F as a function of different values of β for D_a/D_m equals to 0.5.

We present the evolution of $\underline{\underline{D}}$'s diagonal components versus the ratio D_a/D_m in Fig. 3.4 for the case of no binding effect. While the evolution of $\underline{\underline{D}}$'s diagonal components versus the ratio D_a/D_m in Fig. 3.5 is plotted for the case of diffusion with chloride binding effect. We observe that as D_a/D_m increases, the diagonal components also increase. Fig. 3.6 confirm the retarding effect of chloride binding on macroscopic chloride diffusion coefficient.



Figure 3.4: Evolution of \underline{D} 's diagonal components versus the ratio D_a/D_m without binding effect.



Figure 3.5: Evolution of \underline{D} 's diagonal components versus the ratio D_a/D_m with binding effect.



Figure 3.6: Evolution of \underline{D} 's diagonal components versus the ratio D_a/D_m with and without binding effect.

3.4.4 Comparison with Maxwell's equation

In order to check the accuracy of the E-FEM for representing a two-phase material at the meso-scale, D_s is compared with the Maxwell's equation. This equation describes the macroscopic transport property of a binary composite containing spherical inclusions such as :

$$\frac{D_s}{D_m} = \frac{2D_m + D_a + 2V_a(D_a - D_m)}{2D_m + D_a - V_a(D_a - D_m)}$$
(3.39)

where D_s is the single value representing the diffusivity along the three axes, D_m is the diffusivity in the continuous phase (mortar matrix), D_a the diffusivity in the discrete phase (aggregate particles) and V_a is the volume fraction of aggregate particles.

Simulations are carried out on the RVE computed in the previous section. We consider a volumic fraction of aggregate particles corresponding to 11.5, 25.5 and 35.5 %. Fig. 3.7 shows a good aggreement between the numerical results and Maxwell's analytical solution. The computed values tend to be smaller than those of Maxwell's analytical solution. However the error is quite acceptable, it is around



Figure 3.7: Numerical results versus Maxwell's analytical solution for several aggregates volume fraction.

1 %. This trend is also observed in [Abyaneh et al., 2016]. This comparison is established for a meso-scale computation of diffusivity without binding effect. In order to show the efficiency of the E-FEM even with binding effect, we compare in the next section, numerical results with experimental work.

3.4.5 Discussion of model validation with experimental data

A comparison of results of the meso-macro numerical approach and experimental findings is necessary to check the applicability of the numerical method. The comparison is carried out with the experimental work of Sleiman [Sleiman, 2008]. Four tests where established on four concrete samples (B15, B22,B4 and B30). The concrete samples were ground to obtain grains of no more than 5mm. They found that that Langmuir isotherm is well suited to their experimental results for the four types of concrete. The average values of constant α and β were adjusted as a function of the experimental results (Table 3.8).

Concrete type	Isotherm parameters	
concrete type	α	β
B_{15}	2.35 E-03	1.2 E-03
B_{22}	3.40 E-03	1.5 E-03
B_4	4.20 E-03	1.5 E-03
B_{30}	4.80 E-03	1.7 E-03

Table 3.8: Langmuir isotherm parameters for the 4 concretes [Sleiman, 2008].

For each type of concrete, a migration test was performed to obtain the coefficient of diffusion as summarized in Table 3.9.

Concrete type	B_{15}	B_{22}	B_4	B_{30}
Diffusion coefficient (m^2/s)	8.91 E-12	5.20 E-12	2.46 E-12	1.0 E-12

Table 3.9: Diffusion coefficient measured by the migration test [Sleiman, 2008].

Recall that in numerical simulations, we need to represent the individual transport property of aggregate and mortar. The aggregate is considered impermeable in this work. However, in [Sleiman, 2008], the values of coefficient of diffusion of mortar matrix phase are not given. We assume here a mortar matrix diffusion coefficient ranges from 1×10^{-11} to 4×10^{-11} for concrete type B_{30} and B_{15} , respectively, by fitting the numerical macroscopic diffusion coefficient obtained with experimental diffusion coefficient of concrete $-B_{30}$ and B_{15} - found in [Sleiman, 2008]. For the other two types $-B_{22}$ and B_{4^-} , mortar diffusion coefficient are estimated according to the relevant compressive strength (see Fig. 3.8) since the chloride diffusion coefficient and concrete -and consequently mortar- strength are linearly related [Francois and Arliguie, 1999] as shown in Table 3.10. Note that a diffusion coefficient value of $2.34 \times 10^{-11} m^2/s$ for mortar after 30 days sodium chloride solution exposure is used by [Sahmaran, 2007].

Table 3.11 shows that the numerical and experimental results are in accordance.

Moreover, a sensitivity analysis is necessary to investigate the impact of binding

Concrete type	B_{15}	B_{22}	B_4	B_{30}
Compressive Strength MPa	15.4	32	39	51
$D_m (m^2/s)$	4.0 E-11	2.6 E-11	2 E-11	1.0 E-11



 Table 3.10:
 Mortar diffusion coefficient.

Figure 3.8: Diffusion coefficient of mortar for the four type of concrete B_{15} , B_{22} , B_4 and B_{30} .

Concrete type	B_{15}	B_{22}	B_4	B_{30}
Concrete diffusion coefficient (Exp.)	8.91 E-12	5.20 E-12	2.46 E-12	1.0 E-12
Concrete diffusion coefficient (Num.)	8.28 E-12	4.75 E-12	2.70 E-12	1.18 E-12

Table 3.11: Mortar diffusion coefficient.

isotherm parameters α and β on the obtained chloride ions diffusion coefficients. It is firstly supposed that α varies with a fixed β in order to investigate the effect of α . Then we examine the effect of β by varying β and a constant α . The results are shown in Fig. 3.9.

As expected, D_s varies with an increase of α and β . The diffusion coefficient D_s decreases with an increase in α for a given β . While, the increase in β for a fixed α leads to an increase in the diffusion coefficient values. Note that the isotherm parameters for concrete type B_{30} in [Sleiman, 2008] are used here.



Figure 3.9: The effect of increasing of isotherm parameters α and β on computed diffusivity coefficient for D_a/D_m equals to 0.5.

3.5 Conclusion

In this chapter, the FE formulation relies on the Embedded Finite Element method (E-FEM) which introduced in the previous chapter is combined with the homogenization method retained in this chapter to:

- determine Representative Volume Element through numerical homogenization;
- compute macroscopic diffusivity tensors accounting for the microstructure diffusivity properties and whose accuracy is good with respect to analytical or experimental results.

Besides, it is mentioned in chapter 2 that taking into account the microstructure diffusion coefficients and the chloride binding effects are important to consider for a better prediction of the time span until corrosion initiation. Consequently, the numerical implementation of the FE formulation suggested in section (3.2) is addressed to take into account binding capacity effects in macroscopic diffusivity tensors in steady-state conditions.

Comparison with Maxwell's equation and experimental work are performed to show the accuracy of the proposed numerical approach. In next chapter, a mesomacro numerical approach will be presented in order to determine of macroscopic diffusivity tensors in heterogeneous quasi-brittle materials such as concrete with the consideration of cracking.

Chapter 4

Meso-macro approach for the numerical modelling of crack-induced diffusivity in concrete: application to service life estimation

4.1 Introduction

Regarding reinforced concrete civil engineering facilities, some corrosive external agents – sea water and spray, deicing salt or even materials making up concrete (sand or additives), – are susceptible to penetrate cement materials. The most corrosive penetrating agents with regards to concrete are chloride ions. Once cover concrete comes through, chloride ions may reach the reinforced course, erode the passive course, and corrode steel reinforcements when the threshold value is reached [Mehta and Monteiro, 2006]. Consequently, due to deteriorated mechanical performances, the structure durability and service life are affected [Benkemoun et al., 2017b]. In the other hand, the presence of cracks (meso and/or macro) provides a preferential path for the penetration of aggressive agents, accelerating in consequence the degradation of the concrete matrix. Hence, numerical simulations are coupling chloride ions transport and crack formation mechanisms in concrete is a crucial issue. The objective of this chapter is to investigate a meso-macro approach allowing to predict macroscopic diffusivity including the effect of crack-induced diffusivity at mesoscale.

Concerning the coupling, researchers link an equivalent diffusion coefficient D_e to a crack width parameter as shown in chapter one. Herein, the relationship between the crack width and the diffusion coefficient through the crack D_{cr} which proposed in Djerbi et al. [Djerbi et al., 2008] is adopted. In this chapter, the general spirit of the present work is to model concrete at the mesoscale as an heterogeneous quasi-brittle two-phase material based upon the previous work of Benkemoun et al. [Benkemoun et al., 2010]. The main features of this work are presented in part 4.2.

In part 4.3 of this chapter, a meso-macro numerical example is presented to carry out the computation of macroscopic crack-induced diffusivity tensors and also to examine the capability of the approach. In the context of concrete-like materials, both artificial and realistic crack-induced diffusivity are considered.

Finally in part 4.4, we turn to the performance of civil engineering facilities through the investigation of service life prediction.

4.2 Modeling of failure of quasi-brittle materials

Mechanical models suitable for studying concrete fracture can be presented, on mesoor macroscopic scale, by two general families of numerical approaches: continuum approaches and discreet approaches.

continuum models (or smeared approaches)

Several authors used the Continuum Damage Mechanics to simulate the initiation and propagation of microcracks ([Mazars and Pijaudier-Cabot, 1989]; [Oliver et al., 1990], [Kachanov and Montagut, 1986]). Through some of these continuum models, such as damage models, the degradation of the elastic properties of the material have been described by proposing a scalar damage variable (d) through which crack representation is taken into consideration, according to Mazars [Mazars, 1984].

As an aspect of the continuum mechanics, a macrocrack that triggers the appearance of discontinuity in the displacement field even though the structure is regarded as continuous.

Numerical methods, like Finite Element Method, allow to solve the model equations but require an extremely fine mesh size to take into account all heterogeneity aspects, and therefore more time of calculation is required. Besides, these models cannot reflect the variation of the properties of the concrete composites such as -for example-: aggregate, cement paste and ITZ. Accordingly, these models neglect -in general- the heterogeneities of concrete and thus it considered to be a homogenized material.

This type of homogenization is unable to characterize the process of cracking in its entirety, from the initiation of macroscopic cracks in the concrete to the propagation and coalescence of microcracks in addition to the inability to estimate crack opening which play a crucial role. Furthermore, since there is a transition from a state of homogeneous deformations to a state of localized deformations due to the inclusion of strain-softening feature where the fineness of the spatial discretization is required so as the localization band being narrower and narrower, classical Finite Element analyses within a continuum mechanics approach are not able to describe properly the localization of deformation which is of a significant importance and act as a precursor of macro-cracks. Thus, they give mesh-dependent results and required a fine resolution of the crack zone [Tejchman and Bobiński, 2012, Oliver et al., 2002, Crisfield and Wills, 1988].

Other approaches called **Discrete models** (or cohesive approaches) are also used. In these approaches, the failure of structural elements is considered to figure out the nonlinear fracture process. So, the microckrack is represented by two surfaces and assigned as fictitious crack using cohesive interface elements embedded into the mesh.

However, the discrete approach suffers from some numerical problems concerning the characteristic and localization of crack such as crack opening and orientation since the crack discontinuities need to grow along the element edges. Also the fact that the crack is restricted to comply with a predefined path along finite element boundaries leads to unknown crack path a priori.

More disadvantage of these methods could be observed, that is a fine spatial discretization is required to reproduce the complex cracks patterns leading to a high computational costs. Nevertheless, neither of these approaches take into account the fracture processes in detail at the structural level. So it was necessary to develop different numerical technique for the simulation of cracking evolution at the mesoscale. At the mesoscale representation, where the aggregate inclusions are embedded in the cement paste matrix, it is allowed to represent the heterogenities as well as the failure mechanism by introducing a kinematical enhancements of the Finite Element interpolation.

The first enhancement, which already shown in previous chapter, is the weak kinematic enhancement. It is adopted to consider the interfaces between the different phases (sub-domains) included in the concrete composition at mesoscale level by means of a non-adapted meshing strategy [Ortiz et al., 1987, Benkemoun et al., 2010, Roubin et al., 2015b]. By using this strategy, a jump within the strain field is introduced to model the contrast of property in the classical linear elements, leading to the previously defined *weak discontinuity* across the interface incorporated within an element of two different material properties (aggregate and cement matrix for example).

Regarding the representation of crack initiation and propagation, many works in literature seek to introduce Finite Element solutions (commonly indicated as a localization limiter or regularization technique) to get over the problem of the pathological sensitivity of the results to the mesh size due to the softening behavior. Some of these works based on the local approach of continuum mechanics such as the smeared crack approach [Hillerborg et al., 1976] or viscous regularized model [Needleman, 1988]. One of their disadvantages is that it does not provide the orientation of the cracks. Other works based on the non-local continuum approach to failure as it was introduced by [Pijaudier-Cabot and Ba v zant, 1987] or gradient-enhanced models (explicit and implicit gradient models) [Peerlings et al., 2000]. These models exhibit some drawbacks such as the poor description of kinematic fields in the fracture process zone due to the dependency of the non-locality on the distance between neighboring points [Giry et al., 2011].

Two decades ago, more stochastic methods developed to enhance the framework of the standard local continuum model by introducing a *strong discontinuity* kinematics where the crack is explicitly represented as a jump in the displacement fields. This kinematic enhancement is applied within the context of Finite Element formulation either on the nodes of the fractured elements (global) as in Extended Finite Element (X-FEM) [Moës et al., 1999], or on the Gauss points of the fractured elements (local) as in Embedded Finite Element (E-FEM) [Ortiz et al., 1987]. These methods allows to eliminate the use of a characteristic length that is sometimes difficult to identify and represent explicitly crack patterns. According to [Oliver et al., 2006], no obvious differences in results obtained from these two methods (E-FEM and X-FEM). However, a problem associated with the use of X-FEM approach is to increase the size of the stiffness matrix for each new displacement jump inserted, beside the advantage of simple implementation and the reasonable computational time of E-FEM make the authors in [Benkemoun et al., 2010] use E-FEM to perform a numerical fracture model for a cement-like quasi-brittle materials. The E-FEM model used in the above-mentioned work based on a double kinematic enhancement introduced at the element level thanks to the theory of incompatible modes [Ibrahimbegovic and Wilson, 1991b]:

A weak discontinuity [Ortiz et al., 1987] - continuous displacement field and discontinuous strain field - permits to represent the behavior of heterogeneous materials.

A strong discontinuity [Simo et al., 1993] - discontinuous displacement field and unbounded strain field - allows to represent explicitly the micro-cracks that may occur in aggregates or matrix or also in the interface zone. It fulfills the major requirements in terms of capability to provide accurate cracks pattern and cracks opening coupled with material heterogeneity representation in an efficient and mesh independent way. Therefore, many advantages pertaining to the use of this numerical approach might be summarized as:

- Introducing of the non-adaptated meshing process.
- The nodes of its unique homogeneous mesh are placed independently from the morphology of the aggregates.
- A significant amount of computation time is saved since the stiffness matrix size and the total number of unknowns for the global system are identical to the problem without kinematics enhancements.
- The micro-cracks are captured to produce at any of different phases (aggregate or mortar matrix) or so at the interface (debonding).
- The ability to model softening behavior without remeshing is also ensured.

In the present work, we model concrete at the mesoscale as an heterogeneous quasi-brittle two-phase material based upon the previous work of Benkemoun et al. [Benkemoun et al., 2010](cf. [Benkemoun et al., 2017b]). A brief description of this mechanical model will be presented hereafter and for further details on the method of resolution, objectivity with respect to the mesh, its numerical implementation and a number of illustrative examples of the model predictive capabilities, see [Benkemoun et al., 2010, Benkemoun et al., 2012].

4.2.1 Kinematics of weak and strong discontinuity

The first kind of kinematics enhancement (cf. Chapter 2) is introduced for the truss element split into two parts, each having a different Young modulus to represent material heterogeneity by means of weak discontinuity, (see Fig. (4.1)).



Figure 4.1: Split truss element with weak discontinuity and G_1 function [Benkemoun et al., 2012]

Where $G_1^{\oplus/2}$ is a scalar function (first incompatible mode) represents the jump in strain field as:

$$G_{1}^{(0)/(2)} = \begin{cases} G_{1}^{(0)} = -\frac{1}{\theta \ell}, & x \in [0, \theta \ell] \\ \\ G_{1}^{(2)} = -\frac{1}{(1-\theta)\ell}, & x \in [\theta \ell, \ell] \end{cases}$$
(4.1)

According to the "Discrete Strong Discontinuity Approach" [Dias-da Costa et al., 2009], the fracture surface can be simulated to capture the jump in displacement field (strong discontinuity). Then, by the flexibility of the E-FEM and the Incompatible Mode Method, the two-phase bar element was enriched by a second incompatible mode G_2 allowing to incorporate the cracking. The function G_2 makes it possible to capture the cracking either at the interface between phase ① and ② if the element is split into two parts, or in one of the two phases if the element is not split into two parts, (see Fig. (4.2)).



Figure 4.2: Split truss element with strong discontinuity and G_2 function [Benkemoun et al., 2012]

The function G_2 can be written as [Benkemoun et al., 2010]:

$$G_2 = -\frac{1}{\ell} + \delta_{\Gamma} \tag{4.2}$$

where θ in (Equations[4.1,4.2]) represent a non-dimensional function to position the interface between the sub-domains of each split elements, and δ_{Γ} is the Dirac generalized function placed at the interface.

The truss element will show two constitutive models: discrete one over the discontinuity, and elastic continuum model outside the displacement discontinuity. The strong discontinuity is activated in element only when a chosen yield function turn to have zero value, otherwise, the material behavior is still in elastic state. The yield function can be written such as:

$$\Phi = t_{\Gamma} - (\sigma_u - q) \tag{4.3}$$

$$q = \sigma_u \left(1 - exp\left(-[|u|] \frac{\sigma_u}{G_f} \right) \right)$$
(4.4)
where t_{Γ} is the traction vector over the discontinuity, [|u|] is the crack width and q is the stress like variable while the fracture energy G_f represents the area under the t_{Γ} versus [|u|] curve, see (Fig. (4.3(right)).



Figure 4.3: Elastic-quasi-brittle behavior outside the discontinuity (left), at the discontinuity [Benkemoun et al., 2012]

The two enhanced functions $G_1^{(1/2)}$ and G_2 were used in the enhanced strain fields $\tilde{\epsilon}_1$ and $\tilde{\epsilon}_2$ as well as the displacement gradient $\nabla^s u$ as a sum to introduce the total strain field ϵ , such that:

$$\epsilon = \nabla^s u + \tilde{\epsilon}_1 + \tilde{\epsilon}_2 \tag{4.5}$$

The total strain is then written in the context of the EAS (Enhanced Assumed Strain, [Simo and Rifai, 1990]) method for each phase as:

$$\epsilon^{0} = Bd + G_{1}^{0}[|\epsilon|] + G_{2}[|u|], \quad \epsilon^{2} = Bd + G_{1}^{2}[|\epsilon|] + G_{2}[|u|]$$
(4.6)

The strain field (Eq. 4.5) is then introduced in the Hu-Washizu variational formulation [Ibrahimbegovic, 2009] leading to the Finite Element problem. This Finite Element framework will be solved in terms of the displacement field dand the enhanced interpolation parameters $[|\epsilon|]$ and [|u|] for the weak and strong discontinuities, respectively. A local-global solving procedure coupled to a return mapping algorithm [Simo and Hughes, 1997] is considered for the solving process. The crack width values which are computed at the end of each mechanical time step for each finite element, are used to achieve fine coupling at the mesoscale for the purpose of computation of macroscopic mass transport properties as in the next part.

In order to model the macroscopic diffusion properties of a cracked cement-based material according to the former methods, the computation of the displacement jump as the crack width [|u|] is computed firstly by means of the mechanical model shown in part (4.2). The crack width values for each finite element obtained from this model are then used as input data in the chloride ions transport problem through D_m in Eq. 4.7. This leads to a weak coupling at the mesoscale with the mechanical problem. The upscaling method which introduced in chapter Three is then applied with the coefficient D_m in Eq. 4.7.

$$\vec{q}(\vec{x}) = -D_m \cdot \vec{\nabla} c(\vec{x}) \tag{4.7}$$

Numerical examples will be presented in the next part to show two numerical examples: one with an artificially cracked domain, and the other one with mechanically-induced cracked domain.

4.3 Numerical simulation and discussion

We consider a $100 \times 100 \times 100 \ mm^3$ domain as a Representative Volume Element (RVE) for representing concrete-like materials. The weak coupling procedure presented in the previous parts is used to compute the macroscopic diffusivity tensors **D**. The discretization of the cubic domain is realized with 626315 nodes and element

size equals to 1 mm (Fig. (4.4(a)). Remind that in the framework of this thesis, we chose to work with a three-dimensional lattice model (Fig. (4.4(b))). For more details about the three dimensional discretization method and convergence with regard to mesh, the reader is referred to the Ph.D. thesis of Nathan Benkemoun (in French) [Benkemoun, 2010].



(a) cubic domain with an unstructured mesh : 626315 nodes



(b) Lattice finite element mesh

Figure 4.4: 3D diiscretization of cubic homogeneous domain.

4.3.1 Artificially cracked domain: case of of homogeneous material

Several authors has used artificial cracks to assess the effect of crack width on diffusion coefficient [Marsavina et al., 2009, Jin et al., 2010, Wang and Ueda, 2011a, Ishida et al., 2009]. The artificial crack is a single, controllable and simple crack with a constant width where no mechanical simulations are performed and no displacement or forces are imposed. It can be represented by a plane created manually with known position, orientation and width for each finite element existing in the intersecting line of this plane. Herein, in order to create this artificial crack, we generate a plane parametrized by the angle κ in between the second spatial axis Y and the normal to the crack, n_{crack} . Whereas, any value of crack width [|u|] could be imposed, a range



Figure 4.5: $100 \times 100 \times 100 \ mm$ cubic domain with κ equals to 0: the crack (colored in red) is in the plane Y = 50 mm.

of 0 to 500 μm is considered for this work.

To obtain the nine components of the macroscopic diffusivity tensor, the cracking-transport coupling at the mesoscale is achieved for each value of crack width, the upscaling method is applied.

4.3.1.1 Crack located in the horizontal plane

For a $100 \times 100 \times 100 \ mm^3$ cubic domain for the diffusion-governing problem only in mortar matrix, i.e. with no aggregate embedded inside the matrix, and a horizontal artificial crack located at $Y = 50 \ mm$ as shown in Fig. (4.5), the numerical simulations present the effects of crack width on the diffusivity tensor as shown in Fig. (4.6) and (4.7).

These figures illustrate the diagonal and off-diagonal components of macroscopic diffusivity tensor $\underline{\mathbf{D}}$ as a function of the crack width [|u|].

It can be seen in Fig. (4.6) that the diagonal components of the macroscopic diffusivity tensor exhibit an anisotropy behavior since the diagonal components of $\underline{\underline{D}}$ do not increase in value with the same magnitude. It is notable that D_{xx} and D_{zz} , in this case, increase in value with the same magnitude since the

macroscopic crack is positioned in the XZ plane. Whereas it is not the case for D_{yy} , it remains invariant with respect to the value of the crack width, and has a value approximately equal to the one in the sound state, $1.8 \times 10^{-12} m^2/s$ for this numerical example. So, the diagonal components do not evolve in the same way. These results are comparable, with good agreement, with the results of Nilenius et al. [Nilenius et al., 2015]. It is important to state that the general behavior of the diagonal components D_{xx} and D_{zz} of Fig. (4.6) is conditioned by the selected coupling Eq. 4.8 at the mesoscale, such as:

$$\begin{cases} D_{cr}(m^2/s) = 1.8 \times 10^{-12}, & [|u|] < 30 \,\mu m \\ D_{cr}(m^2/s) = 2 \times 10^{-11} [|u|] - 4 \times 10^{-10}, & 30 \,\mu m \le [|u|] \le 80 \,\mu m \\ D_{cr}(m^2/s) = 14 \times 10^{-10}, & [|u|] > 80 \,\mu m \end{cases}$$
(4.8)

Therefore, the diffusivity increases rapidly in value after 30 μm value of crack width.



Figure 4.6: Diagonal components of \underline{D} in relation with the crack width

Fig. (4.7) plots the off-diagonal components of $\underline{\underline{D}}$. Physically speaking, these terms correspond to crossed-interactions. For instance, D_{xy} is the value of $\underline{\underline{D}}$ in the X-direction when a gradient of concentration is applied in the Y-direction. D_{xy} and D_{zy} have the same level of magnitude in terms of value whereas D_{xz} has higher value. This result is consistent regarding the location of the crack, namely in the X - Z plane.



Figure 4.7: Off-diagonal components of \underline{D} in relation with the crack width

According to the effective range of crack width that might alter the diffusivity of cracked medium as stated by Djerbi et al. [Djerbi et al., 2008] of (30 to 80 μm), three values of [|u|] are considered as: 0, 30 and 90 μm to clarify if $\underline{\mathbf{D}}$ is symmetric and positive-denfinite or not. Hereafter, the results of $\underline{\mathbf{D}}$ demonstrate that it is symmetric with real coefficients so it is diagonalizable. Moreover, to say that it is positive-definite, we have to prove that the eigenvalues are positive. The set of eigenvalues for 0, 30 and 90 μm crack width are found to be all positive: $(1.77, 1.77, 1.77)^{0\mu m}$, $(2.52, 1.81, 2.47)^{30\mu m}$ and $(6.79, 1.87, 6.37)^{30\mu m}$ (note that we have skipped $10^{-12} m^2/s$), so $\underline{\underline{\mathbf{D}}}$ is positive-definite.

$$\underline{\underline{D}}^{0\mu m} = \begin{pmatrix} 1.77 & 1.45 \times 10^{-5} & -4.51 \times 10^{-6} \\ 1.45 \times 10^{-5} & 1.77 & 5.48 \times 10^{-6} \\ -4.51 \times 10^{-6} & 5.45 \times 10^{-6} & 1.77 \end{pmatrix} \times 10^{-12} m^2 / s$$

$$\underline{\underline{D}}^{30\mu m} = \begin{pmatrix} 2.51 & 1.79 \times 10^{-3} & -2.30 \times 10^{-2} \\ 1.79 \times 10^{-3} & 1.81 & 2.67 \times 10^{-3} \\ -2.30 \times 10^{-2} & 2.67 \times 10^{-3} & 2.48 \end{pmatrix} \times 10^{-12} m^2/s$$

$$\underline{\underline{D}}^{90\mu m} = \begin{pmatrix} 6.65 & 2.41 \times 10^{-3} & -1.97 \times 10^{-1} \\ 2.41 \times 10^{-3} & 1.87 & 5.27 \times 10^{-3} \\ -1.97 \times 10^{-1} & 5.27 \times 10^{-3} & 6.51 \end{pmatrix} \times 10^{-12} m^2 / s$$

Besides, a comparison between eigenvalues of $\underline{\mathbb{D}}^{0\mu m}$ and of $\underline{\mathbb{D}}^{30\mu m}$ with the diagonal values of $\underline{\mathbb{D}}^{0\mu m}$ and of $\underline{\mathbb{D}}^{30\mu m}$, and their identity generate a valuable conclusion that the off-diagonal terms of $\underline{\mathbb{D}}^{0\mu m}$ and $\underline{\mathbb{D}}^{30\mu m}$ are negligible (terms in $10^{-14} m^2/s$ and $10^{-15} m^2/s$). However, for $\underline{\mathbb{D}}^{90\mu m}$, this conclusion cannot hold. We conclude that when the crack width increases, the macroscopic diffusivity will have important value even in the off-diagonal components and attention must been payed to the induced-anisotropy.

4.3.1.2 Crack rotated at an angle k

To analyze the influence of macro-crack's orientation (rather than its width value), a perfect crack is parametrized by different values of k. Hence, for a crack width [|u|] equals to $90\mu m$, four values of k are studied: 20° , 30° , 60° and 90° as shown in Fig. (4.8)



Figure 4.8: Position of the perfect crack in relation with k.

The results of the numerical computations give the diffusivity tensor $\underline{\underline{D}}_{k}^{90\mu m}$, its eigenvalue $\lambda_{k}^{90\mu m}$ and the corresponding eigenvectors $f_{k}^{90\mu m}$ for crack value equals to $90\mu m$ and different values of k. The same conclusion with the horizontal perfect crack that the diffusivity tensor is symmetric and positive-definite is predicted here. This assures a good agreement with the theoretical work of [Pouya and Courtois, 2002]. Moreover, the diffusivity behavior of diagonal components of the orthogonal planes YX and ZX (i.e. D_{yy} and D_{zz} , respectively) is evolving reciprocally whenever the angle k increases. D_{yy} increases while D_{zz} decreases towards the value of the diffusion coefficient in the mortar matrix, $1.8 \times 10^{-12} m^2/s$. This permutation occurs around k equals to 45° .

Concerning D_{xx} , we can note quasi-symmetric values for κ equals to 0° $(6.65 \times 10^{-12} m^2/s)$ and 90° $(5.99 \times 10^{-12} m^2/s)$ and for κ equals to 30° $(9.15 \times 10^{-12} m^2/s)$ and 60° $(9.12 \times 10^{-12} m^2/s)$ with higher values for κ equals to 30° and 60° . These results can be explained by the evolution of the crack length

value when rotation occurs: for κ equals to 0° and 90°, the crack length value is equal to 100 mm whereas for κ equals to 30° and 60°, it is equal to 115.5 mm. In addition, the higher the crack length value is, the higher the flux passing through the crack is (the length of the crack on the other faces of the cube does not change) and so the higher the value of D_{xx} is.

Furthermore, the anisotropy of the off-diagonal terms is shifted from the plane XZ for $k = 0^{\circ}$ $(D_{xz,0^{\circ}} = -1.97 \times 10^{-13} \ m^2/s)$ to the plane XY for $k = 90^{\circ}$ $(D_{xy,0^{\circ}} = -2.04 \times 10^{-13} \ m^2/s)$.

Also, the resulted values of eigenvectors $f_k^{90\mu m}$ demonstrate an accordance with the corresponding values of crack's inclination angles k such as: 20.03° (for $k = 20^{\circ}$), 29.99° (for $k = 30^{\circ}$) and 60.12° (for $k = 60^{\circ}$).



Figure 4.9: Diagonal components of <u>D</u> in relation with the angle κ for $[|u|] = 90 \ \mu m$

$$\underline{\underline{P}}_{20^{\circ}}^{90\mu m} = \begin{pmatrix} 8.21 & 7.76 \times 10^{-4} & -6.19 \times 10^{-3} \\ 7.79 \times 10^{-4} & 2.25 & 1.14 \\ -6.20 \times 10^{-3} & 1.14 & 4.96 \end{pmatrix} \times 10^{-12} m^2 / s$$
$$\lambda_{20^{\circ}}^{90\mu m} = \begin{pmatrix} 8.21 & 0 & 0 \\ 0 & 5.38 & 0 \\ 0 & 0 & 1.83 \end{pmatrix} \times 10^{-12} m^2 / s$$
$$\underline{f}_{20^{\circ}}^{90\mu m} = \begin{pmatrix} 1.00 & 0.00 \\ 0.00 & 0.34 & 0.94 \\ 0.00 & 0.94 & -0.34 \end{pmatrix}$$

$$\begin{split} \underline{\underline{P}}_{30^{\circ}}^{90\mu m} &= \begin{pmatrix} 9.15 & 4.02 \times 10^{-2} & 6.51 \times 10^{-2} \\ 4.02 \times 10^{-2} & 2.61 & 1.35 \\ 6.51 \times 10^{-2} & 1.35 & 4.17 \end{pmatrix} \times 10^{-12} m^2/s \\ \lambda_{30^{\circ}}^{90\mu m} &= \begin{pmatrix} 9.15 & 0 & 0 \\ 0 & 4.95 & 0 \\ 0 & 0 & 1.83 \end{pmatrix} \times 10^{-12} m^2/s \\ \underline{f}_{30^{\circ}}^{90\mu m} &= \begin{pmatrix} -1.00 & -0.02 & 0.00 \\ -0.01 & 0.50 & 0.87 \\ -0.02 & 0.87 & -0.50 \end{pmatrix} \end{split}$$

$$\underline{\underline{D}}_{60^{\circ}}^{90\mu m} = \begin{pmatrix} 9.12 & -1.22 \times 10^{-2} & -9.24 \times 10^{-3} \\ -1.22 \times 10^{-2} & 4.44 & 1.50 \\ -9.25 \times 10^{-3} & 1.50 & 2.69 \end{pmatrix} \times 10^{-12} m^2 / s$$

$$\lambda_{60^{\circ}}^{90\mu m} = \begin{pmatrix} 9.12 & 0 & 0 \\ 0 & 5.30 & 0 \\ 0 & 0 & 1.83 \end{pmatrix} \times 10^{-12} m^2 / s$$

$$\underline{f}_{60^{\circ}}^{90\mu m} = \begin{pmatrix} 1.00 & 0.00 \\ 0.00 & -0.87 & -0.50 \\ 0.00 & -0.50 & 0.87 \end{pmatrix}$$

$$\underline{\underline{P}}_{90^{\circ}}^{90\mu m} = \begin{pmatrix} 5.99 & -2.04 \times 10^{-1} & -1.95 \times 10^{-3} \\ -2.04 \times 10^{-1} & 6.03 & 3.34 \times 10^{-3} \\ -1.95 \times 10^{-3} & 3.34 \times 10^{-3} & 1.85 \end{pmatrix} \times 10^{-12} m^2 / s$$

$$\lambda_{90^{\circ}}^{90\mu m} = \begin{pmatrix} 6.21 & 0 & 0 \\ 0 & 5.80 & 0 \\ 0 & 0 & 1.85 \end{pmatrix} \times 10^{-12} m^2 / s$$

$$\underline{f}_{90^{\circ}}^{90\mu m} = \begin{pmatrix} 0.67 & -0.74 & 0.00 \\ -0.74 & -0.67 & 0.00 \\ 0.00 & 0.00 & 1.00 \end{pmatrix}$$

4.3.2 Two-phase mechanically-induced cracked domain

The artificial cracks were represented as parallel smooth walls. Until this point, just this feature was considered in the numerical examples -presented previously- as heterogeneity. However, it is obvious to note that artificial crack could not reflect the reality, since real cracks are more tortuous and rough in addition to the existence of embedded inclusions in cement past [Van Belleghem et al., 2016].

In this part of our study, the two heterogeneities are studied where both mechanically-induced cracks and aggregate embedded into a mortar matrix are considered to produce the heterogeneity and then, assess the influence of the combined heterogeneities at mesoscale on the macroscopic diffusivity tensor of concrete as a whole. As a first step of this work, the implementation of mechanical computations is performed as shown briefly in part (4.2) and detailed in [Benkemoun, 2010]. The results of this stage will be presented in the forthcoming part as crack opening values [|u|]. Thereafter, the same simulation procedure - established in part (4.3.1)- is followed to determine the macroscopic diffusivity tensor.



Figure 4.10: $100 \times 100 \times 100$ mm cubic domain with 35 % of spherical aggregates used for the numerical example. The domain has 1878945 DOF for the mechanical problem and 626315 DOF for the diffusion problem.

4.3.2.1 Tensile test simulation

The domain of a cubic shape of $100 \times 100 \times 100 \text{ mm}$ is again considered and discretized. Some differences are regarded here where the heterogeneity is composed, firstly, of macro-cracks, and, secondly, of embedded inclusions in a mortar matrix. Numerical mechanical computations are performed on cement-based composites containing 35% spherical inclusions with two diameter sizes Φ : 4 and 16 mm as shown in Fig. (4.10). The tensile test is simulated here to calculate the values of crack widths. Fig. (4.10) shows two types of lattice elements, obviously, as a set with the same elastic modulus and no strain discontinuity that laying in the mortar or aggregate (colored in blue or white, respectively), and the other type is composed of two materials (mortar and aggregate), simultaneously, that are split by a physical interface and whose strain discontinuity is activated (colored in red).

Thus, a two-phase material and three mechanical property sets are considered at the mesoscale, as in Table 4.1. The numerical simulation of the tensile test applied to the cubic domain is carried out by applying longitudinal or axial displacement along the second spatial axis Y. Fig. (4.11) and (4.12) show the crack evolution for

phase	E (GPa)	$\sigma_u (MPa)$	$G_f (J/m^2)$
mortar matrix	18	3	5
aggregates	127	elastic	elastic
interfaces	-	2	5

Table 4.1: Mesoscale material properties for the numerical simulations

 Φ equals to 4 and 16 mm, respectively.

According to the mechanical elastic parameters of mortar, aggregates and interface in Table 4.1, the aggregate is appear to be stiffer than the other components and, consequently, the micro-crack is initiated around the aggregates in the interface elements corresponding to a weak zone whenever the strong discontinuity is



Figure 4.11: Crack pattern evolution and crack width values for the tensile test with Φ equals to 4 mm.



Figure 4.12: Crack pattern evolution and crack width values for the tensile test with Φ equals to 16 mm.



Figure 4.13: Cut out in the domain showing the macro-crack path tortuosity for Φ equals to 4 (right) and 16 (left) mm

activated(Fig. (4.11(a)) and (4.12(a))). The micro-cracks continue to grow with the increase of macroscale strain to cause cracking coalescence process leading to macro-cracks formation in the direction roughly orthogonal to the imposed displacement and passing around the aggregates and until arriving failure criteria (Fig. (4.11(b), 4.12(b))) and (Fig. (4.11(c), 4.12(c))).

It is easy to draw some conclusions from these results (which have been validated and discussed in [Benkemoun et al., 2010,Benkemoun, 2010,Benkemoun et al., 2012] that no obvious effect of the difference of aggregate size on the maximum crack opening values, otherwise, Fig. (4.11(c), 4.12(c) and 4.13) show that the macrocrack pattern is more tortuous for an aggregate diameter equals to 16 mm and is rotated around the X and Z axis.

Now, it is the appropriate occasion to use the obtained crack opening values in a modified diffusivity-crack width relation to establish the transport-cracking coupling at mesoscale and, then, obtain the macroscopic diffusivity tensor as in the following part.

4.3.2.2 Macroscopic diffusivity tensor computation

As it has been already mentioned, the concrete cubic domain is modeled as twophase material to represent the contrast of property in the classical linear elements. For this reason, the experimental law (Eq. 4.8) of Djerbi et al. [Djerbi et al., 2008] is slightly modified to account for a two-phase material such as:

$$\begin{array}{rcl}
D_{cr}(m^2/s) &=& \theta D_1 + (1-\theta)D_2 & [|u|] < 30\,\mu m \\
D_{cr}(m^2/s) &=& 2 \times 10^{-11}[|u|] - 4 \times 10^{-10} & 30\,\mu m \le [|u|] \le 80\,\mu m \\
D_{cr}(m^2/s) &=& 14 \times 10^{-10} & [|u|] > 80\,\mu m
\end{array}$$
(4.9)

where D_1 and D_2 are the diffusion coefficient in the mortar matrix and in the aggregates, respectively, and θ is the spatial position of the interface in an element lays in both mortar and aggregate as shown in Fig. (4.1). Note that when an element is either in mortar or in aggregate, D_1 is equal to D_2 and still having the corresponding mesoscopic diffusion property and the experimental original expression of Djerbi et al. [Djerbi et al., 2008] is again employed.

The aggregate is set as relatively impermeable is considered in several numerical works [Du et al., 2015, Savija et al., 2013, Sun et al., 2011], and has a diffusion coefficient equals to $D_2 = D_1/100$. While, mortar diffusion coefficient value is $D_1 =$ $1.8 \times 10^{-12} m^2/s$, and accordingly, interface coefficient diffusion determined as in Table 4.2.

phase	$D_m (m^2/s)$	
mortar matrix	Eq. (4.9): $D_1 = D_2 = 1.8 \times 10^{-12}$	
aggregates	$D_2 = 1.8 \times 10^{-14}$	
interfaces	Eq. (4.9): $D_1 = 1.8 \times 10^{-12}$ and $D_2 = 1.8 \times 10^{-14}$	

 Table 4.2:
 Mesoscale material diffusion coefficient for the numerical simulations

Fig. (4.14) shows the diagonal components of the macroscopic diffusivity tensor

as a function of the maximum crack width. These components have been computed from the upscaling procedure performed from the mesoscale transport-cracking coupling for the last time step corresponding to a maximum crack opening equals to 0.52 mm for Φ equals to 4 mm and 0.53 mm for Φ equals to 16 mm. By looking again to Fig. (4.12(a)), we can understand the slight increase in the values of the diagonal components of the macroscopic diffusivity tensor that appears in Fig. (4.14). This slight increase comes from the initiation of micro-cracks around the stiff aggregate at early stage of crack formation for a maximum crack width ranging from 0 to 0.25 mm.

One more region can be observed regarding this plot starting from a threshold value around 0.25 mm where the mechanical simulations have exhibited the peak in the stress versus strain curve and micro-crack coalescence process leads to a macrocrack (see Fig. (4.11(b) and 4.12(b))). As shown in Fig. (4.14), a significant increase in the values of the diagonal components of the macroscopic diffusivity tensor is observed. As a conclusion, we see that the macroscopic diffusivity tensor integrates the evolution from diffuse cracks to localized crack. Such feature comes as a by-product result of the meso-macro analysis.

The analysis is also applied to assess the effects of aggregate size on diffusivity. Taking another look to Fig. (4.14) conducts us to conclude that rising of aggregate size used from 4 to 16 mm leads to an increase of 8 %, 16.4 % and 4.7 % between $D_{xx,4mm}$ and $D_{xx,16mm}$, $D_{yy,4mm}$ and $D_{yy,16mm}$, $D_{zz,4mm}$ and $D_{zz,16mm}$, respectively.

The longer and more tortuous crack path for Φ equals to 16 mm is the reason of the increase in the diagonal components values obtained as clearly observed in 4.13. This tortuosity leads to a rotation of the macro-crack around the X and Z axis with



Figure 4.14: Diagonal components in relation with the maximum crack width and the aggregates size a rotation more important around the X axis.

Basing on the results of this analysis, a service life prediction is established using the diagonal components values of the macroscopic diffusivity tensor to assess the impact of cracks-induced diffusivity on durability of concrete structure. In the following part, a simple physical model is employed to obtain the corrosion initiation time depending on the macroscopic diffusivity.

4.4 Service-life estimation

Regarding performance and durability of Civil engineering facilities, service life estimation is a crucial issue. Service life of concrete structures might be defined as the period through which a structure achieves its performance requirements without needing unexpected repair services until damage increase arrives at an impermissible level. It is very often linked to the ability of concrete to prevent the penetration of aggressive agents in its porous network. According to the widely acceptable model of [Tuutti, 1982], the service life of a structure is divided into initiation t_{\circ} and propagation t_p period. The initiation of corrosion begins when the concentration of chloride around the reinforcement exceeds the threshold chloride concentration (C_{th}) . Consequently it is related to the penetration of chloride ions to steel surface level as shown in Fig. (4.15), This



Figure 4.15: Service life of concrete structure subjected to corrosion [Tuutti, 1982, Adiyastuti, 2005].

initiation time can be described mathematically by a simple physical model proposed by Bažant [Bazant, 1979], as:

$$t_{\circ} = f(C_s, C_{th}, D, L) = \frac{d_c^2}{12D \left[1 - \sqrt{\frac{C_{th}}{C_s}}\right]^2}$$
(4.10)

Where d_c is the concrete cover (m), D the diffusion coefficient of chloride ions (m^2/sec) , and C_s the surface concentration (kg/m^3) . The propagation period t_p is considered as the time between initiation and corrosion-induced cracking where the concrete cover begins to be deteriorated by cracking and spalling because of the increase of corrosion products. Furthermore, Bažant [Bazant, 1979] introduce

another model to identify the corrosion propagation time t_p as in Eq. (4.11):

$$t_p = \rho_{cor} \frac{d_s}{s} \frac{\Delta d_s}{j_r}, \Delta d_s = 4f_t \frac{L}{d_s} \delta_{pp}$$
(4.11)

Here, ρ_{cor} is the corrosion product density, d_s is the diameter of the bar, s the space between steel bars, j_r the rust production rate, δ_{pp} the bar hole flexibility. Therefore, the critical time at which corrosion starts to induce cracks is ([Bazant, 1979]):

$$t_{cr} = t_{\circ} + t_p \tag{4.12}$$

Service life estimation, to be as realistic as possible, has to integrate at least the main degradation factor of concrete structures into service, i.e. cracking. Consequently, we can note here the contribution of the proposed upscaling method in the context of service life estimation: both cracking and microstructure are integrated as a by-product result of the meso-macro analysis into the macroscopic diffusivity tensor components that can be plugged in service life data, t_{\circ} and t_{p} . We propose to use the aforementioned model of Bažant to estimate the service life t_{cr} , by figuring out the initiation period of corrosion as an indication adopted by some of previous study as ([Zhang and Lounis, 2006], [Liu, 1996] and [Kwon et al., 2009]).

The variation of the predicted service life (initiation and propagation phases) associated with the values of crack width is investigated with the following parameters used by [Liang et al., 2009]: concrete cover $d_c = 40(mm)$, $\frac{C_{th}}{C_s} = 0.32$, $j_r = 1.5 \times 10^{-15} (g/m^2.s)$, the corrosion product density $\rho_{cor} = 3600 (kg/m^3)$, the space of the steel bar and s = 0.1(m). The corrosion-induced cracks time t_{cr} according to Bažant models are illustrated in Fig. (4.16 and 4.17) in relation to crack

width values and aggregates size.

We note that t_{cr} is affected by the crack width in an anisotropic manner: around 0.5 mm of crack opening value, t_{cr} is almost divided by two in comparison with a sound concrete for the xx and zz components and whatever the size of the aggregates is. For the yy component, t_{cr} is smaller for D = 4 mm than for D = 16 mm. This observation has to be correlated with the smaller diffusivity computed for D = 4mm in comparison with D = 16 mm (see Fig. (4.14).



Figure 4.16: Time to induce cracking related to crack width variation for 4 mm aggregate size

From these results it can be clearly observed that a direct link between cracking phenomenon and heterogeneities at a fine scale can be related to service life models at the macro scale which might be considered as useful information for engineering analysis and design requirments.

4.5 Conclusions

We have presented in this chapter a meso-macro numerical approach accounting for crack- induced diffusivity in heterogeneous quasi-brittle materials such as concrete.



Figure 4.17: Time to induce cracking related to crack width variation for 16 mm aggregate size

The meso-structure is based on a two-phase 3D representation of heterogeneous materials where the aggregates are embedded within a mortar matrix.

The previously introduced weak and strong discontinuities have been considered as well to account for the heterogeneity and crack evolution, respectively. The crack opening values obtained from the mechanical analysis for each Finite Element at each time step have been used in the experimental law of Djerbi et al [Djerbi et al., 2008] to realize the cracking-diffusion coupling at mesoscale. The information from the mesoscale to the macroscale are upscaled by means of an homogenisation method based on the work of Pouya and Courtois [Pouya and Courtois, 2002].

The macroscopic diffusivity tensor is the mean goal of this chapter which aim to obtain material property at macroscale from a fine-scale information in combination with the mechanical damage effect.

As a conclusion of this chapter, we stress the interest of the proposed upscaling method coupled to a mesoscale analysis to outline the following features:

- The macroscopic diffusivity tensors exhibit anisotropic behavior.
- The increase in the values of the diagonal components of the macroscopic diffusivity tensor starts at a crack opening value threshold of 0.25 mm.
- The higher value of the aggregates diameter corresponding to a more tortuous macro-crack path - gives a higher value of the diagonal components of the macroscopic diffusivity tensor.
- The macroscopic diffusivity tensors can be used to improve the estimation of the service-life of reinforced concrete structures.

Chapter 5

General conclusions and

perspectives

A Finite Element formulation to model chloride ion diffusion numerically in mesoscale concrete, taking account of chloride binding capacity, is described in the first part of this study. This formulation is based on the Embedded Finite Element method (E-FEM) which enables meshes that may not correspond to the physical interface, such as the aggregate/mortar matrix for mesoscale concrete, to be used without losing the accuracy of the classic finite element approach. This is achieved by introducing a weak discontinuity in the chloride concentration field to take into account the heterogeneous nature of concrete-like materials with no mesh adaptation. The validity of the FE formulation, carried out in the finite element code FEAP [Taylor, 2008], is confirmed for analytical solutions for a 1D semi-infinite two-phase material. Firstly, it is used to examine the impact of (1) the diffusion coefficient values of the aggregate and mortar matrix and (2) chloride binding on changes in concentration profiles. Once the evolution of concentration profiles for different time intervals is known, the corrosion initiation time t_{corr} can be assessed. Thus, t_{corr} is found to:

- decrease when the effects of chloride binding capacity are taken into account;
- increase when the ratio of diffusion coefficients $D_{matrix}/D_{aggregate}$ increases.

Clearly, in order to predict better t_{corr} , it is important to consider explicitly the diffusion coefficients of aggregate and cement matrix with the consideration of chloride binding effects. This is crucial for an accurate estimation of the service life of a structure.

In the next part, 3D simulations are presented. The homogenization method used here in combination with the E-FEM is shown to be effective for:

- revealing the Representative Volume Element by numerical homogenization;
- calculating the macroscopic diffusivity tensors, which represent the microstructure diffusivity properties and are accurate regarding Maxwells equation.

In addition, the FE formulation that takes account of the binding capacity effects on macroscopic diffusivity tensors in steady-state conditions is implemented numerically. Comparison with experimental work is also performed to show the applicability of the proposed numerical approach regarding the case of binding effect.

It is shown that, in order to rely on chloride ion transport models, regardless of their complexity, the main factor causing the degradation of concrete structures in service -cracking- must be taken into account.

In the second part of this study, a meso-macro numerical approach that considers the diffusivity due to cracking in heterogeneous quasi-brittle materials such as concrete is described. In addition to the weak discontinuity, a strong discontinuity is also added to take into account micro-cracking. In fact, this strong discontinuity is the main feature of this work with regard to the mesoscale coupling which provides the crack opening values for each finite element at each time step. Thus, at the mesoscale, each crack is considered to be a pathway for an oriented flow whose diffusion coefficient obeys the experimental law of Djerbi et al. [Djerbi et al., 2008].

The same homogenization method used in the first part is employed in this model resulting in the calculation of macroscopic diffusivity tensors considering fine scale information.

Although the fine scale models are simple (elastic-brittle behavior for the mechanical model and Ficks law in fully saturated conditions for the mass transport model), more complex features are included in these macroscopic diffusivity tensors as a secondary result of the two-scale analysis. The major features considered are:

- the induced-anisotropy as shown in chapter 4. Although the mass transport model does not rely on an anisotropic model, macroscopic diffusivity tensors behave anisotropically.
- the development of diffuse cracks in the bulk to localized macro-cracks as described in Section 4.2.2. Fig. [4.14] shows that an increase in the diagonal component values of the macroscopic diffusivity tensor is generated above the crack-opening threshold value of 0.25 mm (which is the peak in the stress versus strain curve for the mechanical simulations).
- the tortuosity of the macro-crack path. Fig. [4.12] illustrates that the macrocrack becomes more tortuous as the aggregate diameter increases, which can be explained by the fact that the macro-cracks are localized around the elastic aggregates. This phenomenon directly impacts the diagonal component values

of the macroscopic diffusivity tensor as shown in Fig. [4.14]. Thus, a larger aggregate diameter, leading to a more tortuous macro-crack pathway, entails a higher diagonal component value of the macroscopic diffusivity tensor. Note that there is no tortuosity parameter in the mechanical model. The tortuosity of the macro-crack pathway is once again included in the macroscopic diffusivity tensor properties as a secondary result of the meso-macro analysis.

In addition, the macroscopic diffusivity tensors found to be usable to improve the estimation of the service-life of concrete reinforced structures, including the effect of the cracking and the internal meso structure.

However, more experimental work is recommended to validate the numerical results of this study.

As perspective for future work, we propose to extend the present model to consider the case of cracked-unsaturated concrete at the mesoscale. Furthermore, the mechanical model can be also extended to account for the mechanism of shrinkage cracking. For this case, the shrinkage-induced crack width can be consider as the crucial information for the computation of mass transport within cracks and thus to achieve fine couplings at the mesoscale. To improve this study, other perspectives are required. We can note:

- To consider a multi-species modelling of chloride ingress which is more realistic than Fick's law.
- To carry out an experimental study dealing with the coupling of cracking that induced by bending, compression,...., and chloride ingress. For this aim, the tidal simulation available in the laboratory could be used.

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Thèse de Doctorat

Mohammed Naji HAMMOOD A Meso-Macro Numerical Approach for Chloride Diffusivity Modeling Taking into Account Chloride Binding and Crack Evolution in Concrete

Résumé

La pénétration des ions chlorure est la principale cause de la dégradation des structures en béton, par corrosion des armatures, entraînant un impact sévère sur leur durabilité et leur durée de vie. La pénétration de ces agents agressifs pourrait être favorisée davantage par la présence de fissures. Dans cet thèse, nous avons utilisé la méthode des éléments finis (EF) pour résoudre l'équation de la loi de Fick couplée à la capacité de fixation d'ions chlorure afin de modéliser la diffusion des ions chlorure à l'échelle mésoscopique. Dans un premier temps, nous avons considéré une représentation 3D d'un matériau, sain, hétérogène biphasé (comme le béton) ou les inclusions (granulats) sont noyées dans une matrice de mortier. Le problème des interfaces (inclusion/matrice) a été résolu en utilisant la méthode E-FEM (Embedded Finit Element Method). Au niveau de ces interfaces, nous avons introduit une discontinuité faible du champ de concentration de chlorures. Une approche d'homogénéisation par moyennes spatiales se basant sur les travaux de Pouya est également utilisée pour prédire les tenseurs de diffusivité macroscopiques des matériaux biphasiques. La comparaison avec l'équation de Maxwell et des résultats expérimentaux a été réalisée pour montrer la précision de l'approche numérique proposée. Dans un second temps, l'approche méso-macro est représentée pour introduire un modèle numérique capable de fournir des informations macroscopiques (tenseur de diffusion moyen) intégrant le niveau d'ouverture de fissure, le chemin de fissuration et l'hétérogénéité des matériaux quasi fragiles tels que les matériaux cimentaires (béton, mortier,). Dans ce cas, des points clés du processus de fissuration comme l'évolution d'une fissuration répartie vers une fissuration localisée (macro-fissure(s)), la tortuosité de la fissure et son anisotropie sont intégrées naturellement dans la diffusivité macroscopique. En fin, le tenseur défini est ensuite utilisé afin d'estimer la durée de vie des structures en béton, y compris l'effet de l'endommagement et de la mésostructure interne

Abstract

The penetration of chloride ions has an essential responsibility in the degradation of concrete structures caused by reinforcement corrosion leading to a severe impact on the durability and service life of concrete structures. The problem becomes more critical with the existence of cracking which accelerate the penetration of chloride ions into concrete cover. In this work, the FE formulation for the numerical modelling of chloride ions diffusion accounting for chloride binding capacity in mesoscale concrete is introduced. The mesostructure is based on a twophase 3D representation of heterogeneous materials, such as concrete, where stiff aggregates are embedded into a mortar matrix. For this purpose, we turn to the Embedded Finite Element Method (E-FEM). This is performed by introducing a weak discontinuity in the chloride concentration field for finite elements where the physical interface is present. Numerical spatial homogenization experiments based on Pouya's works are also performed on 3D mesostructures to compute macroscopic diffusivity tensors accounting for two-phase material. Comparison with Maxwell's equation and experimental results are carried out to show the accuracy of the proposed numerical approach. Finally, the meso-macro approach is presented to introduce a numerical model capable of providing macroscopic information (mean diffusivity tensor) integrating the level of crack opening, crack path and heterogeneity of materials in quasi-brittle concrete. The mesoscale coupling with the mass transport part is based on Fick's Law with a modified diffusion coefficient taking into account crack opening and aggregates. The macroscopic diffusivity tensor integrates more complex features such as the cracking evolution process, tortuosity of the crack's path, inducedanisotropy and presence of aggregates. The defined tensor is used afterwards in order to estimate the service-life of concrete structures, including the effect of the cracking and the internal mesostructure.

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