APPROXIMATION BY THE FINITE ELEMENT METHOD TO THE PREFERENTIAL CHLORIDE DIFFUSION THROUGH INTERFACIAL TRANSITION ZONE IN CONCRETE

Y.A. VILLAGRÁN ZACCARDI[†] [‡] and V.L. TAUS[†] [‡]

† *Área Tecnología del Hormigón, LEMIT, CICPBA, 1900 La Plata, Argentina. hormigones@lemit.gov.ar*
‡ *CONICET, Argentina*

Abstract— **Chloride diffusion in concrete is a main aspect of reinforced concrete durability. It defines the time required for reinforcement corrosion in marine structures. Concrete porosity is one of the main concerns involved in the process of chloride ingress from the environment into concrete. However, concrete can hardly be considered homogeneous in the meso-level. Natural coarse aggregates are usually less porous than the cementitious matrix, whereas interfacial zones between aggregates and the matrix are the most porous phase in concrete. This aspect is difficult to be experimentally studied, as very small samples need to be collected and analyzed. As an approach, the diffusion process can be simulated with the Finite Element Method (FEM). In this paper, chloride diffusion into concrete is simulated with a 2D FEM model, distinguishing three phases with different porosities in the material. Interfacial zone is identified as preferential path for chloride ingress. The study reveals a significant influence of particle inclusion on the chloride diffusion into concrete, and the effect of the particle shape.**

Keywords— **Chloride, Concrete, Diffusion, Interfacial Transition Zone, FEM.**

I. INTRODUCTION

Reinforced concrete plays a key role in the construction industry. It is the most used material for structural purposes. Durability is one of its advantages over other materials, especially metallic materials. However, some durability issues may appear with time in reinforced concrete. Chloride is present in the marine environment, and it ingress into concrete by absorption, permeability and diffusion. When the threshold chloride content is reached at the reinforcement depth level, steel depassivates and corrodes. Cover concrete eventually cracks due to the pressure of expanding oxides. Then, the process speeds up with steel more exposed to the environment. Chloride ingress into concrete is usually associated with diffusion through concrete pore structure. This is a simplification of the multimechanistic process.

Concrete is basically constituted by fine aggregate (sand), coarse aggregate (stone), Portland cement, and water. Hardened concrete is an artificial porous rock. A fraction of mixing water reacts with cement in the hardening process, whereas pores are due to evaporation of non-reacting water. In the meso-level, hardened concrete may be divided in three phases: aggregate, interfacial transition zone (ITZ), and matrix. Each one has a certain porosity level. The properties of aggregates are those of the rock of which they are constituted. ITZ and matrix properties depend on the proportions of constituents in concrete, curing conditions, age, admixtures, cement content, type of aggregates, and others. ITZ in concrete is originated, to a large extent, by the formation of water-filled spaces around aggregate particles in the fresh mix. ITZ is thus characterized by an increased porosity relative to the matrix phase and most of the rocks commonly used as aggregate. Therefore, it is the weakest zone for the transport of aggressive agents.

In general, concrete is treated as a homogeneous medium as regards chloride ingress (Andrade, 1993; Martin-Perez *et al.*, 2001; Poulsen and Mejlbro, 2006) and characterized by a single diffusion coefficient. However, diffusion cannot be homogeneous in concrete when considered as a multiphased material.

The influence of ITZ on chloride diffusion into concrete may be affected by geometrical parameters such as the particle shape of aggregates. Proximity is the main factor that defines the type of aggregate used in concrete making. Some very common types are crushed stone (e.g., granite, basalt, sandstone), and rounded gravel (from a river or seashore). The respective angular or rounded particle shapes define two cases for the observation of chloride diffusion in three-phased concrete.

Natural aggregates usually have a much lower porosity than that of the matrix or ITZ. Therefore, transport through the aggregate phase is generally negligible in concrete. Assuming this simplification, coarse aggregate particles act as obstacles for the chloride ingress process. However, these obstacles are surrounded by the most porous phase in concrete, ITZ, and transport is favored on the aggregate boundaries.

Aggregate particles in the matrix have two effects. The addition of solid obstacles increases tortuosity, and chloride ions must move around these particles. On the other hand, the formation of ITZ facilitates ion mobility. The relative importance of these two opposite effects will define the overall transport affectation by the aggregate inclusion. In this analysis, parameters such as aggregate absorption, maximum aggregate size, flatness and elongation, emerge as important issues.

Preferential diffusion through ITZ in concrete is hardly demonstrable by experimental work. Very small samples need to be collected and analyzed to describe chloride transport through the meso-structure of concrete. Here, the Finite Element Method (FEM) is noticed as a useful tool to approximate the phenomenon.

An adequate mesh would permit distinguishing the three phases that have to be considered. Calculating the ingress rate is allowed by the time evolution of the approximation.

Previous investigations have used the FEM to model 2D chloride diffusion into saturated concrete (Shin and Kim, 2002; Suwito *et al.*, 2006; Wang *et al.*, 2007; Guzmán Gutiérrez, 2010). Concrete has been considered as a homogeneous material in these studies, however; and the potential influences of the ITZ and particle size and shape were disregarded. Though Zeng (2007) modeled a two-phased concrete (aggregate and matrix), the main focus was on the blocking effect of particles, and the very relevant influence of the ITZ was not investigated.

II. DIFFUSION IN CONCRETE

Diffusion is due to a concentration gradient. Flow occurs from the zone with higher concentration to the zone with minor concentration. Diffusion through the solid phase of concrete is negligible at room temperature. The particular case of chloride transport takes place in the voids of concrete filled with liquid. As a result, aspects regarding saturation degree and pore connectivity must be taken into account. These aspects are included in the diffusion coefficient value.

Fick's Second Law describes the diffusant transport under non-stationary conditions (Eq. 1). One-dimensional form of this law is shown in Eq. (2), where *D* is not a function of position or time. It is widely used to model chloride diffusion in saturated concrete.

$$\frac{\partial C}{\partial t} = \nabla^2 (D \cdot C) \tag{1}$$

$$\frac{\partial C}{\partial t} = D . \frac{\partial^2 C}{\partial x^2}, \tag{2}$$

where C is the chloride content at depth x and time t, ∇^2 is the Laplace operator, and D is the diffusion coefficient.

The most widespread solution to Eq. (2) is Eq. (3), considering conditions in Eq. (4) and Eq. (5). Derived for a semi-infinite medium, Eq. (3) allows determining the concentration as a function of position and time.

$$C(x,t) = C_0 \left(1 - erf\left[\frac{x}{2\sqrt{Dt}} \right] \right) \tag{3}$$

$$C = 0, x > 0 \wedge t = 0 \tag{4}$$

$$C = C_0, x = 0 \wedge t > 0 \tag{5}$$

Knowing the value for D implies the possibility of estimating the initial time when a threshold chloride quantity reaches reinforcement and depassivates it.

III. EXPERIMENTAL

Preferential transport through ITZ was analyzed by tests on concrete probes. Concrete slices were polished and exposed on one of their edges to a 1% blue methylene solution, for 72 hours. The solution ascended by capillarity through concrete pore structure. Hence, the most porous phases in concrete were visually detected considering the longer distance reached by the colored solution due to the higher transport rate.

Concrete with a water/cement ratio of 0.50 and granitic crushed stone as coarse aggregate was examined in the sorptivity test. Figure 1 shows the obtained results. Aggregate particles are clearly enclosed by the solution. Thus, ITZ is identified as the most porous phase in concrete.

The progress of the colored front was in this case only by capillarity. Although concrete was not fully saturated, sorptivity and diffusion are processes both dependent on porosity, and their ingress front depths are ruled by proportionality to the square root of time. The methodology presented was useful for qualitative identification of the phases in concrete.

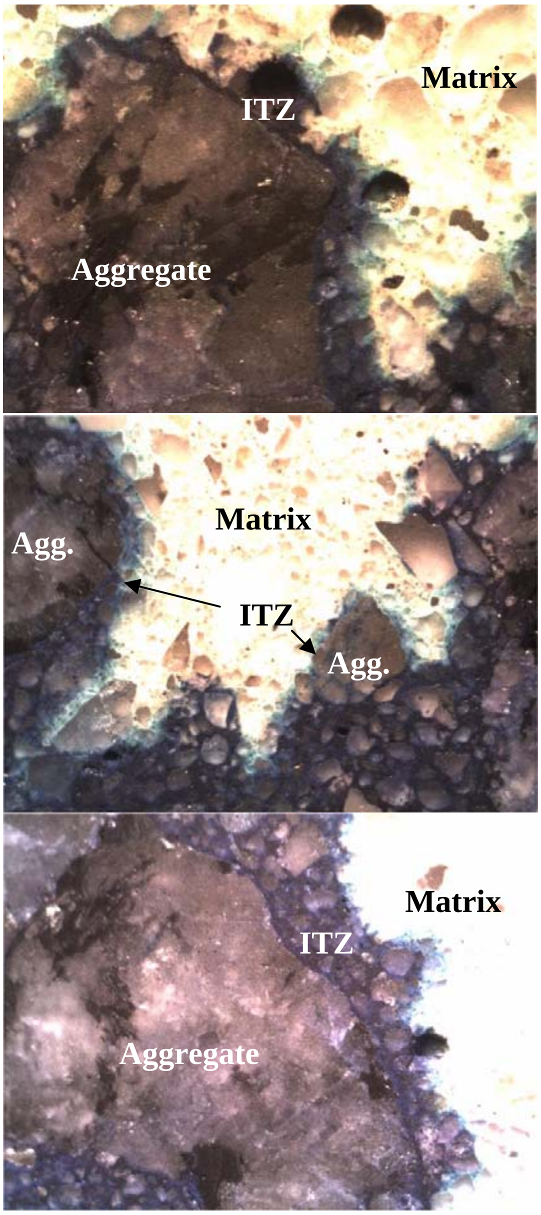

Figure 1. Details of concrete sorptivity of 1% blue methylene solution.

IV. FEM IMPLEMENTATION

Following the finite element method (FEM), the non-linear diffusion (Eq. 1) was turned into a set of algebraic equations (Kwon and Bang, 1997), to model chloride diffusion into concrete in a domain $\Omega \subset \Re^2$. Values for D and C_o were according to literature (Poulsen and Mejlbro, 1996; Traversa, 2001; Nilsson *et al.*, 1996). They are consistent with saturated concrete immersed in salt water. Chloride binding and diffusivity time evolution were disregarded. Both influences may be significant; however, some discrepancies on how they must be considered persist in literature (Poulsen and Mejlbro, 1996; Traversa, 2001; Nilsson *et al.*, 1996; Glass and Buenfeld, 2000; Mangat and Molloy, 1994; Stanish and Thomas, 2001). Therefore, it was preferred them not to be included.

The balance represented in Eq. (1) is rearranged, and, following the method of weighted residuals, the differential equation is multiplied with an arbitrary weight function w, and integrated over $\Omega \in \Re^2$. Integration by parts of the Laplacian leads to the weak formulation, Eq. (2), in the Sobolev space $H^1(\Omega)$: For all $w \in H^1(\Omega)$ and times $t \in (0,T)$, holds Eq. (6).

$$-\int_{\Omega}\left(\frac{\partial w}{\partial x}\frac{\partial u}{\partial x}+\frac{\partial w}{\partial y}\frac{\partial u}{\partial y}\right)d\Omega-\int_{\Omega} w\frac{du}{dt}d\Omega=0 \qquad (6)$$

$$\forall w \,/\, w(0)=0 \qquad (7)$$

The shape functions used were those for linear triangular elements, and satisfied the conditions of Eq. (8) and Eq. (9). Here, δ_{ij} is the Kronecker delta.

$$H_i(x_j,y_j)=\delta_{ij} \qquad (8)$$

$$\sum_{i=1}^{3} H_i = 1 \qquad (9)$$

The triangular mesh generated presented a progressive refinement through aggregate boundaries. Isoparametric elements were optimized by aiming their shape to isosceles triangles. Unknowns were approximated in terms of the nodal values. For n elements, discretization can be expressed as follows,

$$\bigcup_{e=1}^{n}\Omega_e=\Omega \qquad (10)$$

$$\bigcap_{e=1}^{n}\Omega_e=\varnothing \qquad (11)$$

Using the Galerkin method, the shape functions H_i are used as weighting functions (Eq. 12). The variable $u=u(x,y,t)$ is interpolated within a finite element using shape functions H_i (Eq. 13).

$$w=H_i \qquad (12)$$

$$u(x,y,t)=\sum_{i=1}^{3}u_i(t)H_i(x,y) \qquad (13)$$

The final matrix equation for Eq. 1 becomes Eq. (14).

$$M\cdot\{\dot{u}\}^t + K\cdot\{u\}^t = \{F\}^t \qquad (14)$$

The time derivative is solved using the backward finite difference method. Equation (14) is turned into Eq. (15). Time derivative in backward differences is Eq. (16).

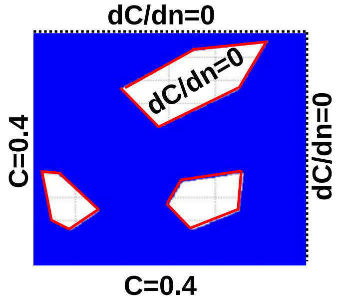

Figure 2. Boundary conditions for three-phased concrete.

The procedure results in Eq. 17 (Kwon and Bang, 1997).

$$M\cdot\{\dot{u}\}^{t+\Delta t} + K\cdot\{u\}^{t+\Delta t} = \{F\}^{t+\Delta t} \qquad (15)$$

$$\{\dot{u}\}^{t+\Delta t} = \frac{\{u\}^{t+\Delta t}-\{u\}^{t}}{\Delta t} \qquad (16)$$

$$([M]+\Delta t[K])\{u\}^{t+\Delta t} = \Delta t\{F\}^{t+\Delta t} + [M]\{u\}^{t} \qquad (17)$$

The modeling of 2D diffusion was carried out on an area corresponding to an edge of a structural element. Concrete was considered as composite material, where mortar matrix contains coarse aggregate particles. Both effects of the inclusion of particles and of their shape were analyzed.

The inclusion of aggregates into the matrix generates weaknesses in the ITZ, and it boosts transport by diffusion. A first approach considered the following boundary conditions (Fig. 2):

a) $\partial C/\partial n=0$, at right and upper sides, and at aggregate-mortar interface. The upper and right sides are imposed and unreal, as the material is further continuous. They are parallel to the transport direction, and therefore, there is no flow of matter through them. This is true when considering a homogeneous matrix on both sides of these limits, which results in the presented boundary condition.

b) C=0.4 at left and bottom sides, in coincidence with the exposed concrete surface to the chloride solution.

With this scheme a differentiated diffusion coefficient may be considered for each of the three phases. An established proportionality between these coefficients let reduce the parameter just to multiples of D. Diffusivity through aggregates may be disregarded, as it is minimal compared to the transport in the matrix and through ITZ.

This proposition leads to a refinement of the mesh through ITZ, and it allows excluding aggregate particles from the meshing.

V. RESULTS AND ANALYSIS

Figure 3 presents the mesh obtained for crushed stone. Figure 4 shows the sequence of chloride ingress modeling. From the upper left graph to the bottom right graph increasing time steps of 15 months are shown making a total period of 10 years in the figure.

It resulted in a clear lack of influence of the ITZ. This lack of preferential diffusion through ITZ results from a misjudgment. Figure 4 shows a behavior that suggests a homogeneous matrix, without transport through the aggregate mass. The particles of aggregate act as blocking elements to the ion flow, and they generate a gathering of chloride on their outer side. Only when the gradient reaches a sufficient level, chloride will continue to ingress and enclose the particles.

Nevertheless, ITZ is not differentiated from the matrix. The reason for this to occur may be the boundary condition on the boundary of aggregate particles, $\partial C/\partial n=0$, which implies the requirement of the shape function developing a steep slope next to the ITZ. As this zone was modeled with a single line of linear triangular elements, one or two of their nodes are restricted by the boundary condition, which results in the rest of nodes being indirectly restricted as well. Using multiple lines to model ITZ would imply a higher refinement because ITZ is a very narrow limit, with a consequent increase in the computational cost. Alternative boundary conditions on ITZ may be used.

Boundary conditions simulating chloride transport rate were introduced at the ITZ. The first point in the ITZ to be reached by the diffusant ingress front, named ‘eye’, was identified for each particle to be the one nearest to the surface (Fig. 5). Here, both angular shaped and rounded particles were modeled. Crushed stone produce an ITZ of better quality due to its angularity and roughness in comparison with gravel, which has a lower adherence to the matrix. Two meshes were employed to consider each of these aggregate shapes (Figs. 3 and 6).

In practice, the ‘eye’ is not necessarily the nearest to the surface. Concrete is heterogeneous at this scale of study, and different defects such as macropores or segregation may determine its location. Concrete isotropy is also controversial when considering phenomena such as exudation, curing treatment, and carbonation. Additionally, the corner of the domain has to be carefully analyzed if an aggregate particle is eventually located there, which may be reached by the diffusant front at an ‘eye’ determined by the bidirectional ingress.

From the ‘eye’, preferential linear diffusion takes place through particle boundaries, at higher rate than in the matrix. The ratio between the transport rate in the ITZ and in the matrix would be mainly determined by the ratio between porosities of each phase (being affected by factors such as water/cement ratio, maximum aggregate size, fine content, mineral admixture contents, curing, cement type, concrete age, compaction degree, cracking). A ratio of 10 between diffusivities in the ITZ and the matrix was considered as an approach to a conventional concrete. However, this value needs to be experimentally corroborated.

Crushed stone particles were shaped as polygons, and the linear diffusion was computed through the segments of these polygons, using Eq. (3) with the ‘eye’ as diffusant source.

Figure 7 shows the sequence of chloride ingress along the time in concrete with crushed stone. A differentiation between transport in the ITZ and in the matrix is observed. The mean penetration of the ingress front is highly different from the obtained in Fig. 4. Results for rounded gravel are shown in Fig. 8.

This last methodology involves the dependency of the diffusant content along the boundary of aggregate particles on its relative distance from the eye, and on the chloride content at that point. This is a limitation when ingress from a different direction results in higher chloride content somewhere in the ITZ. Although distances from other diffusant sources remain constant, the gradients in those directions may grow with time.

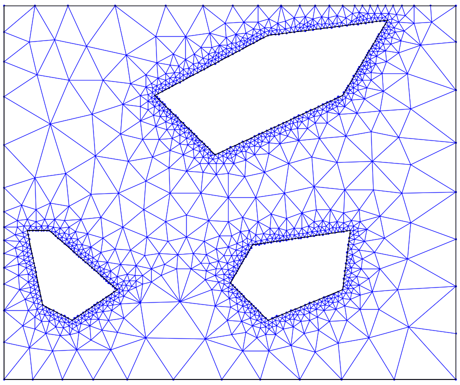

Figure 3: Mesh for concrete with crushed stone.

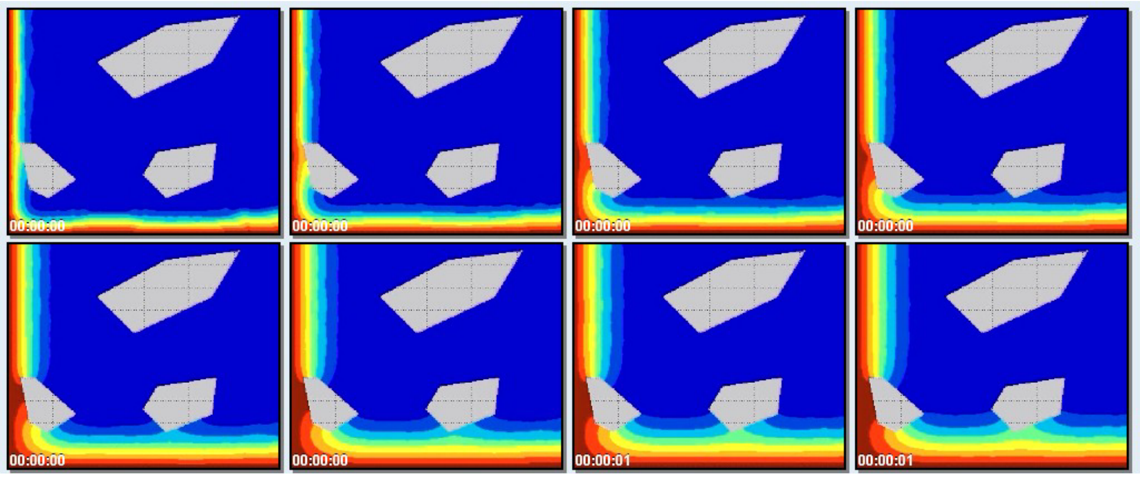

Figure 4: Sequence of chloride ingress into three-phased concrete with boundary conditions in the ITZ.

Therefore, the first main direction of ingress may be surpassed due to higher increasing rate of the chloride content in a different point. In addition, linear diffusion simulated in the ITZ by the boundary conditions will eventually become stationary, and it will not grow with time from that moment forward. This is possible before diffusion is stationary throughout the matrix. If so, ITZ will be conditioned by contents lower than those that are necessarily influenced by the flow from the matrix with higher chloride content. Consequently, the presented methodology seems adequate for a limited period of time after exposure.

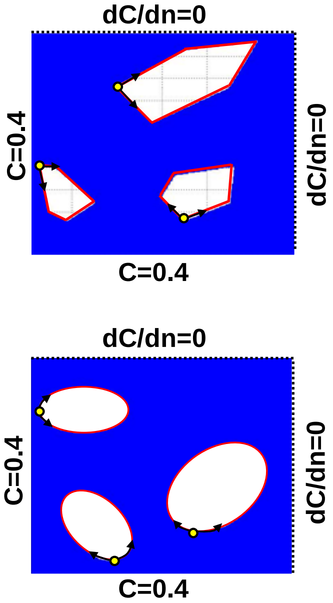

Figure 5: Preferential diffusion through the ITZ, in concrete with: (a) crushed stone (b) rounded gravel

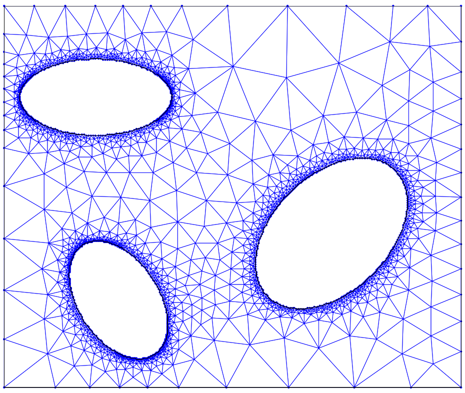

Figure 6: Mesh for concrete with rounded gravel.

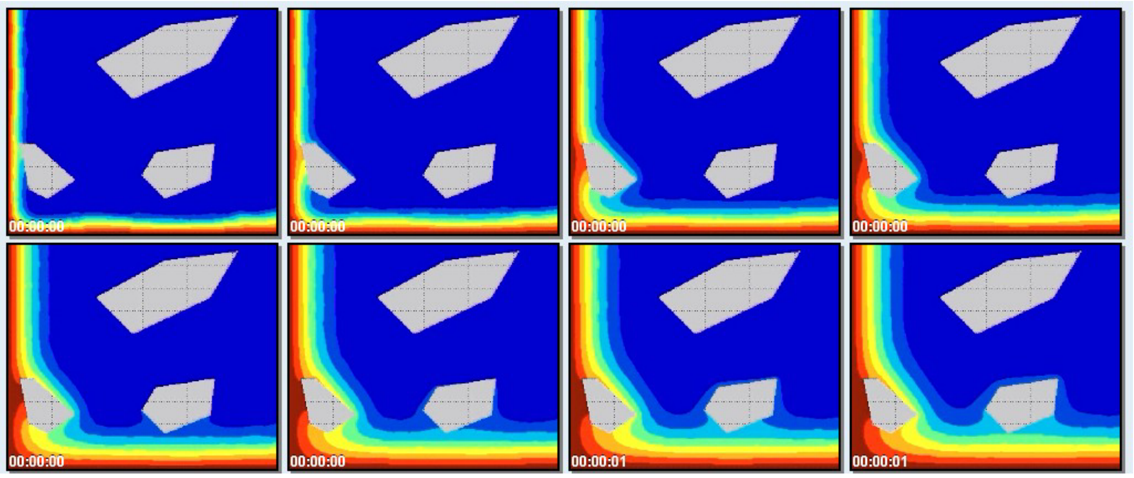

Figure 7: Sequence of chloride diffusion into concrete with crushed stone, considering preferential ingress through ITZ at the boundary of particles.

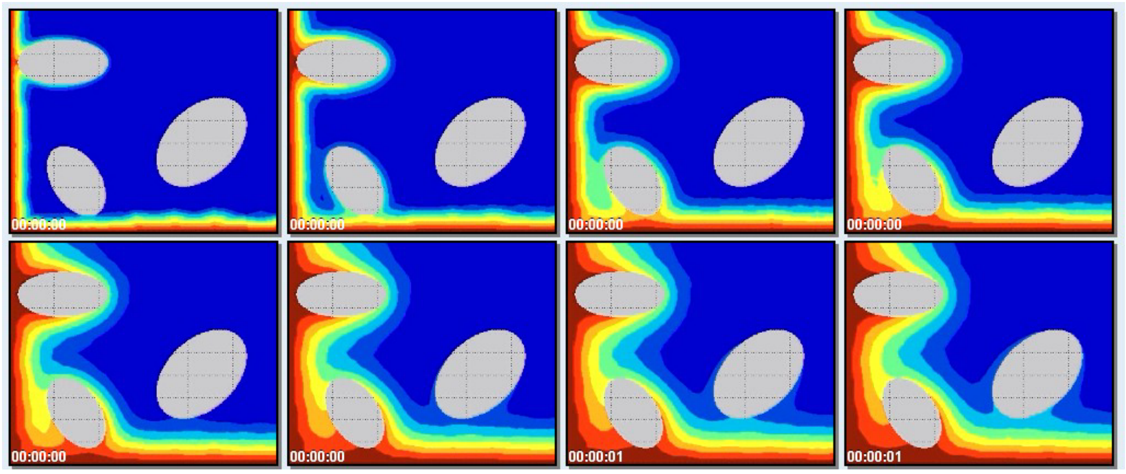

Figure 8: Sequence of chloride diffusion into concrete with rounded gravel, considering preferential ingress through ITZ at the boundary of particles.

Regarding meshing quality, the following indexes have been calculated: (a) the ratio of the diameter of the circle inscribed to the longer side of the element; (b) the double of the ratio of the diameter of the circle inscribed in the element to that one of the circle circumscribing the element; (c) the ratio of the acutest inner angle in the element to $\pi/3$. Average values for these indexes were close or higher than 0.80 in the meshes presented.

From comparison between Figs. 7 and 8, it can be seen that ingress fronts progress differently. Rounded particles have a lower perimeter/area ratio, which means a lower perimeter than angular particles with the same maximum aggregate size. This influence seems lower as particles are more elongated. The results show that if only the particle shape is considered, a rounded particle close to the surface causes a weaker zone than an angular particle. This effect would be incremented by the lower quality of the ITZ that rounded gravel develops in comparison with crushed stone.

VI. CONCLUSIONS

A 2D FEM model to predict the influence of coarse aggregate on the chloride ingress into concrete was presented.

The effect of particles was considered both as regards tortuosity increase, and ITZ formation. Different results were obtained according to the particle shape, due to the perimeter/area ratio and the angulosity.

The approach shows a big potential for further research on the inclusion of the effect of the saturation degree and the binding capacity of concrete.

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