GENERATION OF NUMERICAL AGGREGATES IN A CONCRETE RVE: INFLUENCE OF INCLUSIONS GEOMETRIC PARAMETERS ON THE MACROSCOPIC TRANSPORT PROPERTIES OF THE MATERIAL

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Abstract
This study aims at generating numerical 3D Representative Volume Elements (RVE) of concrete so as to study the effects of the granular inclusions shape on the macroscopic kinetics of transport phenomena. Two phases are considered in the RVE: a matrix of mortar and inclusions corresponding to the concrete coarse aggregates. The choice of a mesoscopic modeling for the mortar matrix is based on the need to obtain numerical structures of reasonable size. In particular, the Interfacial Transition Zones (ITZs) are ignored, as this hypothesis seems acceptable for large aggregates. This study is applied to simple modeling approaches of reactive transport phenomena: drying, and atmospheric carbonation. The purpose is to quantify the influence of the aggregate shape on the kinetics of macroscopic transfer and the isolines for some physical variables representative of the reactive transport problems: saturation degree and calcite concentrations. Basic aggregates shapes are studied (spheres, cubes), as well as more complex and random geometries (Voronoi particles) which are supposed to be more representative of real aggregates. The effects of ‘non-isotropic’ shapes (oblate and prolate ones) are also investigated.

1. INTRODUCTION
Concrete may be viewed as a heterogeneous material made up of a matrix of hardened cement paste in which are distributed granular particles of various size and shape. The durability of this material is known to be highly dependent on its transfer properties. In particular, the evolution of the saturation degree due to drying and of the microstructure influences significantly the migration of aggressive substances (e.g., carbon dioxide,
chlorides...) from the environment. As the main mechanisms of transport and chemical degradation occur in the cement paste phase, it is of great interest to quantify the effects of the granular particles on these mechanisms. Especially, the effects of their shape and their relative spatial arrangement and orientation are expected to be important with regards to the variability of the saturation degree and chemical degradations. These factors may in turn influence significantly other deleterious phenomena as steel corrosion.

In this study, we investigate by means of 3D finite element (FE) numerical simulations the effects of inclusive particles of different shapes on both drying and atmospheric carbonation. For this purpose, several 3D microstructures with various particle shapes are generated. Drying and carbonation are modelled very simply by one and two coupled mass balance equations, respectively. The numerical results are analyzed and compared in terms of time evolution of the mean value and coefficient of variation of the depth at which some characteristic quantities (saturation degree for drying, calcite concentration for carbonation) reach a given value (80% for $S_r$ and 0.01 for the calcite volume fraction).

2. 3D MICROSTRUCTURE GENERATION

The 3D microstructures are generated by randomly dispersing particles of given shape in a box. The minimal distance between inclusions is prescribed by the user. The purpose being to analyze the effects of the particle shape on drying and carbonation phenomena, we propose to limit our study to microstructures with one surface exposed to environment. This requirement is fulfilled by imposing a non-overlapping condition between granular particles and the surfaces of the box, then by cutting all the surfaces (i.e. reducing the dimensions of the box) except for the exposed surface. This implies that the initial box dimensions should be greater than the final ones. The influence of the particles shape is investigated through different samples generated with spherical and cubic inclusions, and with more complex shapes obtained by extracting particles from a classical Voronoi particle assemblage. Several cases of prolate (long) and oblate (flat) shapes have also been analyzed. These shapes are simply obtained by applying a scale transformation on the Voronoi particles in one (prolate particles) or two (oblate particles) directions. To limit the computational time, the number of inclusions is imposed to be 470 whatever the particle shape, corresponding to an initial volume fraction of 0.297. It is noteworthy that due to the random placement of particles and of the subsequent cutting procedure, the effective final volume fraction is microstructure-dependent and ranges from 0.326 to 0.34 (see Table 1 for further details). The Figure 1 shows the particles arrangement obtained for the spherical (left), Voronoi (center) and prolate Voronoi (right) shapes. The face exposed to drying and carbonation appears in the foreground left of the microstructures, while the other faces are cut.

A software developed in python language and making use of the geometry module of the integration platform for numerical simulations Salome [1] automatically generates the samples (see e.g. [2] for further details). This module provides a list of functions for working with computer-aided design (CAD) models and can correct them to be consistent with meshing algorithms. Once the geometry of the structure is performed, a mesh can be generated via Salome. We used the automatic meshing softwares BLSURF and GHS3D developed by Distene to compute the surface meshes and the volume ones. BLSURF is a mesh generator for surfaces composed of parametric patches, conforming to a prescribed size map. GHS3D is a meshing software component which creates automatically tetrahedral
meshes out of closed triangular surface meshes. The number of tetrahedra in the different meshes generated is listed in Table 1. Due to the curvature of the sphere surfaces, the element number needed for their discretization is greater than for the other shapes which exhibit plane surfaces. Note that the conditions imposed for creating the meshes, i.e., a maximum element size of 2 mm, is similar for all microstructures. The number of elements in the matrix which will be used for the computations (the particle phase is not included in the calculations) ranges between about 280000 and 500000.

![Figure 1: Microstructures generated with spheres (left), Voronoi particles (center) and prolate Voronoi particles (right)](image)

<table>
<thead>
<tr>
<th></th>
<th>Spheres</th>
<th>Cubes</th>
<th>Voronoi (long) 1.5</th>
<th>Prolate (flat) 2</th>
<th>Oblate (long) 3</th>
<th>Oblate (flat) 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume fraction of inclusion</td>
<td>32.66</td>
<td>33.45</td>
<td>33.57</td>
<td>34.01</td>
<td>32.99</td>
<td>32.59</td>
</tr>
<tr>
<td>Total number of tetrahedra</td>
<td>736410</td>
<td>417882</td>
<td>474241</td>
<td>457124</td>
<td>452346</td>
<td>537719</td>
</tr>
<tr>
<td>tetrahedra number in the matrix</td>
<td>498549</td>
<td>277457</td>
<td>327587</td>
<td>309821</td>
<td>311302</td>
<td>378278</td>
</tr>
</tbody>
</table>

3. **DRYING AND CARBONATION MODEL**

The numerical model proposed for coupled concrete drying and carbonation is a simplified version of the model initially proposed in [3], and then modified in [4]. This model is governed by the coupled mass conservation equations written for water (1) and carbon dioxide (2). Equation (1) is driven by water pressure \( P_l \). \( K(\phi) \) is the intrinsic permeability coefficient, depending on the porosity \( \phi \). The evolution of the permeability with the porosity is ruled by the relations given in [5], as well as the relative permeability for liquid \( k_r(S_r) \) and
the relation between the saturation degree $S_r$ and the porosity. Equation (2) is driven by the partial pressure of carbon dioxide in the gaseous phase $P_{c, f(\phi, S_r)}$ and $D_{c}$ are the reduction factor and diffusion coefficient of carbon dioxide, respectively, so that the product of both can be considered as the effective diffusion coefficient of carbon dioxide through the porous material. The reduction factor is expressed in [6].

$$\frac{\partial \left( \rho \phi S_r \right)}{\partial t} = \nabla \left[ K(\phi) \frac{P_{c, f(\phi, S_r)}}{\eta} k_{r}(S_r) \nabla (P_{c}) \right] + W_{H_{2}O} \tag{1}$$

$$\frac{\partial \left( (1 - S_r) \phi P_{c} \right)}{\partial t} = \nabla \left[ f(\phi, S_r) D_{c} \nabla (P_{c}) \right] + W_{CO_{2}} \tag{2}$$

The coupling between equations (1) and (2) appears in the rates of water formation $W_{H_{2}O}$ and carbon dioxide dissolution $W_{CO_{2}}$ expressed in Equations (3) and (4), respectively; $N_{cal}$ is the molar formation rate of calcite; $N_{por}$ and $N_{CSH}$ denote the molar dissolution rate of portlandite and of other hydrates, respectively. The coefficient $\alpha_{H_{2}O}$ stands for the average number of moles of water obtained from the dissolution of one mole of ‘various’ hydrates. The expressions of these molar formation rates appear in Equations (5-7), where $k_{H}$ is the Henry constant for carbon dioxide, and $k_{sl}(S_r)$ is a function introduced in [7] to describe at a macroscopic scale the influence of the saturation degree drop on the chemical reaction processes (reduction of water and of contact surfaces between water and cement hydrates). The functions $k_{p}(V_{p})$ and $k_{CSH}(C_{cal})$ are kinetic coefficients depending on the volume fraction of portlandite and the concentration of calcium in solid phase, respectively.

$$W_{H_{2}O} = M_{H_{2}O} \phi S_r \left( \dot{N}_{por} + \alpha_{H_{2}O} \dot{N}_{CSH} \right) \tag{3}$$

$$W_{CO_{2}} = -RT \phi S_r \dot{N}_{cal} \tag{4}$$

$$\dot{N}_{cal} = \dot{N}_{por} + \dot{N}_{CSH} \tag{5}$$

$$\dot{N}_{por} = k_{sl}(S_r) k_{sl} \dot{N}_{CSH} \left( C_{cal} \right) P_{c} \tag{6}$$

$$\dot{N}_{CSH} = k_{sl}(S_r) k_{sl} \dot{N}_{CSH} \left( C_{cal} \right) P_{c} \tag{7}$$

The kinetic coefficient for the portlandite dissolution is assumed to evolve as proposed in [8]: the calcite is supposed to form a growing layer surrounding the cementitious hydrates. The kinetics of the calcite precipitation due to the portlandite dissolution is driven by the diffusive phenomena through the calcite layer. The kinetic coefficient for the other hydrates is assumed to evolve linearly with the calcium concentration [4]. The main simplification of this numerical model in comparison to the previous versions [3]-[4] is the calcium concentration resolution. In the previous versions of the model, an equation of reactive diffusion was solved, coupled with water and carbon dioxide mass balance equations. In the present approach, no diffusion of calcium is considered (meaning that the calcium dissolved from portlandite and other hydrates is locally transformed into calcite), but replaced by a local chemical equilibrium of calcium.
This model was implemented in the FE code Cast3M [9], and solved with a fully implicit numerical scheme. The accuracy of this model was tested by comparison with both numerical results for atmospheric carbonation and experimental results for accelerated carbonation as presented in [4]. The model turned out to be relevant and suitable for numerical simulations on the large meshes considered in this study (up to 500000 elements). The drying simulations have been performed with an initially saturated material exposed to a saturation degree equal to 65% on one face, and we focus on the depth in the material where the saturation degree reaches the arbitrary value of 80%. The issue of drying is equivalent to a simple diffusive problem: no source term appears in the Equation (1) in this case, and the porosity of the material and the transport properties do not evolve during the simulation. On the contrary, the carbonation issue is more complex, due to the coupling between the two phenomena (transport of water and of carbon dioxide) through the source terms of Equations (1) and (2). The simulations have been achieved with conditions of atmospheric carbonation (pressure of carbon dioxide equal to 35 Pa on the external face). The carbonation depth shall be considered for our concern and for the sake of comparison as the point in the material where the calcite formation reaches the value of 0.01 m$^3$ of calcite per m$^3$ of porous material.

4. RESULTS AND DISCUSSION

Before the analysis of the drying and carbonation depths, let us observe on Figure 2 the evolution of the (mean) volume fraction of coarse aggregates in the material (a depth equal to 0 stands for the face exposed to the external environment). These evolutions are obtained from an averaging procedure performed on the surfaces parallel to the exposed one. The volume fraction starts at 0, since this face is not overlapped by aggregates. It increases to a maximum value for a depth equal to about 10 mm, and then reaches a value of about 35% with variations ranging between approximately 32 and 38%. It is noteworthy that the volume proportion profile is similar for every shape tested. This means that the differences in the saturation degree profiles and the calcite profiles that might be observed for the various coarse aggregates shapes shall not be explained by significant disparities in the volume fraction profiles.

![Figure 2: Evolution of the volume fraction of aggregates in the material](image-url)
One can observe on Figures 3 and 5 the mean values for the drying and carbonation depths as defined previously, respectively. In both cases, these mean values are very close whatever the aggregate shapes, except for the oblate (flat) Voronoi aggregates with a shape factor of 3 (meaning very anisotropic shapes). For this latter case, the mean values are slightly inferior to the values obtained for other shapes.

The second observation derives from Figures 4 and 6, showing the coefficient of variation of the drying and carbonation depths along these depths. It appears that the coefficient of variation remains very close for all isotropic shapes, and even for those that are slightly anisotropic (cubes, spheres and Voronoi particles with shape factor less than 2). The coefficient of variation reaches an extreme value in the region near the maximal volume fraction of coarse aggregates (see Figure 2). This phenomenon might be explained by the fact that in the first millimetres, there is essentially mortar and few aggregates. Therefore, little difference can be observed between all cases investigated. The volume fraction of aggregate is then dependent, for its maximal value, on the aggregate shape: the more ‘anisotropic’ the aggregate, the more disturbed the geometry, and thus, the transport processes. Therefore, the coefficient of variation for the drying/carbonation depth reaches its maximal value in the same zone as the volume fraction of aggregates, and this may also explain why the coefficient of variation
variation is more important for widely anisotropic aggregate shapes. It is worth mentioning
that for such anisotropic shapes, the particles seem to exhibit a preferential orientation, i.e.
‘parallel’ to the exposed surface, as may be seen on Figure 1 right in the case of elongated
aggregates. The corresponding spatial arrangement in the region close to the external face is
then probably different from that deeper in the material. This point will be investigated in
more details in a subsequent contribution. For further drying/carbonation depths, the
coefficient of variation decreases until a value that we infer to be asymptotic.

Figure 5: Mean value for the carbonation depth (corresponding to a calcite concentration
equal to 0.01)

Figure 6: Evolution of the coefficient of variability for the carbonation depth

The main conclusion of this study would be that the kinetics of the transport processes we
considered, as well as their local variability, depend essentially on the volume fraction of
aggregates and would not, or hardly, depend on the aggregates shapes, provided the
considered shapes remain ‘rather’ isotropic. For largely anisotropic aggregate shapes (but
with a still isotropic spatial distribution and orientation of the inclusions), the transfer
processes appear to be slightly slower, and the local spatial variability slightly more
important. These conclusions should however be considered with caution, as the Interfacial
Transition Zones have not been taken into account in the modelling approach and only the
course aggregates have been investigated. Therefore, it appears that for a macroscopic
modelling approach involving transport processes (such as drying or carbonation), the influence of the aggregate shapes could be neglected, and the aggregates could be chosen of the shapes that seems more convenient for the numerical simulation (easier to build the geometry with spherical inclusions, less Finite Elements in the mesh with cubic inclusions, etc.). Let us keep in mind also that only transport processes have been taken into consideration, and the conclusions we drew from this study cannot be directly extended to mechanical behaviour.

5. CONCLUSIONS

This study investigates the effects of the shape of coarse aggregates on the macroscopic drying and carbonation phenomena in concrete by means of 3D FE simulations. Numerical microstructures are generated involving 470 granular particles randomly placed and oriented. Different aggregate shapes are considered: ‘isotropic’ ones (spherical, cubic, Voronoi particles) and ‘anisotropic’ ones (prolate and oblate Voronoi particles with various scale coefficients). The numerical results obtained with the different microstructures are compared in terms of time evolution of the depth at which the saturation reaches 80% and the calcite volume fraction is equal to 0.01 for the drying and carbonation problems, respectively. It is shown that all configurations provide very similar results, except for the case with oblate (flat) Voronoi aggregates obtained with a scale transform coefficient of 3. In this case the depths are slightly lower, indicating globally smaller transport properties, and the coefficients of variation are higher in the region near the face exposed to the environment. These discrepancies may be explained by slightly different spatial arrangement and orientation of the aggregates in this zone. Overall, the main conclusion is that for the drying and carbonation phenomena considered in this study, the influence of the coarse aggregate shapes appears negligible (very low for flat shapes) compared to their volume fractions. The same analysis will be carried out in a future work with microstructures containing steel reinforcement rebars near the exposed surface.

REFERENCES