MULTI-SCALE COUPLED PORE STRUCTURE MODELING APPROACH TO CONCRETE; SKETCH OF STRATEGY AND METHODOLOGY, AND EXAMPLES

P. Stroeven, H. He, Z. Guo and M. Stroeven

Faculty of Civil Engineering and Geosciences, Delft University of Technology, the Netherlands

Abstract
Pore structure in concrete gives route for the transport of harmful substance from outside into concrete, which deduce deterioration of concrete or corrosion of steel reinforcement. Therefore, it plays an important role of characterization of pore structure in durability prediction of concrete. Proper physical computer simulation approach, like concurrent algorithm-based systems, renders an economic alternative for material character and properties prediction instead of time-consuming experimental investigation. In this paper, different level of pore structure characterization approaches has been investigated. Furthermore, the applicability of two concurrent algorithm-based simulation systems, i.e. SPACE and HADES, in pore structure assessment of concrete is analyzed. A method coupled micro-level and meso-level of pore structure characterization in model concrete is revealed. Both successful application results examples and future work will be discussed in this paper.

1. INTRODUCTION

Durability receives major attention nowadays when designing concrete structures, particularly when positioned in harsh environment. Transport of harmful substances through pore structure can lead to deterioration of concrete or to corrosion of reinforcement. As the main transport route, the geometric and topological characteristics of pore structure are thus of crucial importance and will be focal elements in this paper. They are studied in model concretes, produced by concurrent algorithm-based SPACE and HADES systems.

Generally, macro and capillary pores are considered effective elements in the transport system, and thus important for concrete durability. Specifically, shape, size and contiguity of pore channels in the Interfacial Transition Zone (ITZ) are of relevance for transport phenomena. Morphological changes of pore structure are assessed by quantitative image analysis, by stereological estimation, and by mathematical morphology methods. This contribution considers elaborating and transforming the new concepts and methodology developed in the previous studies of the authors into a more complete and operational system.
These earlier studies on micro-level indicated that connected porosity is concentrated in very narrow zones around aggregate grains. The local permeability of bulk cement paste on the micro-structural level can be neglected. Further, this implies that a study of pore percolation in larger concrete units should concentrate on (near) particle contiguity in the aggregate skeleton. The surface to surface nearest neighbor distribution is therefore an important characteristic of this skeleton. When global concrete durability is at issue, the meso-level should therefore be selected. This leads to the so-called multi-scale modeling approach.

In this paper, two concurrent algorithm-based discrete simulation systems, i.e., SPACE and HADES, were employed for generating the aggregate structure, encompassing spherical as well irregularly shaped particles. ITZ layers will be added to the surfaces of aggregate grains to complete the cherry-pit model. Attention will focus in first place on modeling aspects.

2. PROBLEM DESCRIPTION

Hydraulic properties are of global or engineering nature, so define properties of a representative volume element (RVE) of the material. For a discussion of the underlying fundamental concept of stochastic heterogeneity, see [1]. The same holds for geometric and topological features of material structure that serve to build a model of the material. The more primitive approach to modeling the pore system is schematizing it into mono-size tubes with cylindrical shape and arranged in parallel mode. This concept is obviously so remote from reality that improvements indeed have been realized. Network modeling is nowadays considered a more realistic approach when based on major geometric and topological features of the real material, the realcrete. Nevertheless, it concerns still a highly idealized version of the complex tortuous multi-connected pore structure in the realcrete. For a survey of some earlier contributions and more present-day approaches, see [2,3].

The strategy we will outline in this paper is based on recent work conducted at Delft University of Technology in cooperation with Chinese scholars [4-7]. In the present framework we will outline the main lines, promoting the general understanding of this basically quite fundamental research approach to durability problems. For details, reference will be given to the relevant literature. This strategy will also form the scope for forthcoming porosimetry research on compucrete produced by concurrent algorithm-based DECS systems developed at Delft University of Technology (SPACE and HADES [8,9]).

In the freshly compacted concrete, the cement paste is mainly pocketed between two or more aggregate (gravel and sand) grains. As is well-known from other fields, the cement particles will pile up against the rigid aggregate grain surface in a non-uniform way. This leads to wall effects extending over a distance equal to maximum grain size, as analytically described by Zheng [10]. The grading of the cement reveals as a result a gradient over this boundary zone, with disproportionately finer particles adjacent to the surface of the aggregate grain. This size segregation phenomenon is significantly stimulated by vibration during compacting, whereby the coarser particles can move away much further from the aggregate grain’s, provided workability conditions are proper (Fig. 1). This is due to the Brazil Nut Effect (BNE), a phenomenon recently getting considerable attention in science. [11].

Size segregation in the fresh state of the cement paste will inevitably sort effect on the hydration process and, as a consequence, on the accompanying evolution of pore de-percolation. It was found that under practical conditions for high strength concrete, the fraction of percolated porosity at ultimate degree of hydration was restricted to a very narrow
zone inside the ITZ adjacent to the aggregate grain surface (Fig. 2). The percolated fraction of porosity encompasses tubes that are not uniformly distributed along this narrow zone. Instead, they reveal concrete’s patchy nature [12,13]. Hence, even with significant overlap of neighboring ITZs, percolation in the overlap zones will be incomplete, supposedly giving rise to the so-called ink-bottle effect, in agreement with similar suggestions in [14]. This is fundamentally different from the common concept of large pores at the nodes that are connected by narrow throats [2]. So, we can conclude here that the pore topology of concrete will be governed by aggregate packing on meso-level, whereas the pore de-percolation process is controlled by cement paste pocketed between aggregate grain surfaces at micro-level. Due the configuration-sensitive nature of such pore features, DECS should not be relying on a random generator, as many of the common systems do, but should involve a particle interaction algorithm, so fall in the category of concurrent-algorithm based systems [15].

![Fig. 1. Section images of fresh model cement paste parallel to rigid surface revealing grading gradient due to compaction vibration. $d_{\text{max}}$ is maximum grain size.](image)

$3 \, \mu m = 0.4d_{\text{max}}$  
$8 \, \mu m = d_{\text{max}}$  
$35 \, \mu m = 4.1d_{\text{max}}$

**Fig. 1.** Section images of fresh model cement paste parallel to rigid surface revealing grading gradient due to compaction vibration. $d_{\text{max}}$ is maximum grain size.

![Fig. 2. (left) Gradient structures of porosity and (right) of the fraction of percolated porosity in high strength concrete at 75% degree of hydration (DOH) in model cement (particle range 1-20 $\mu m$, w/c=0.3 Blaine number=300 $m^2/kg$) [4].](image)
3. VIRTUAL REALITY CONCRETE: COMPUCRETE

3.1. Particle packing
Available physical computer simulation methods for forming granular packing of hard particles can be placed in two distinctive groups. Systems based on sequential random (particle) addition (SRA) were developed, using a random generator algorithm. Such systems generate more evenly distributed particles than met in practice, and neglect as a consequence phenomena such as flocculation, agglomeration and clustering. The particles are placed rather than packed. Therefore, it is impossible to achieve total packing densities relevant for concrete aggregate by such systems. Concurrent algorithm-based simulation (CAS) systems were developed starting from SRA procedures, avoiding such limitations. These systems can realistically simulate the particulate structure. Different strategies for incorporating particle interaction are followed, but in dynamic systems a so-called dynamic stage is doing this. The SPACE system, developed at Delft University of Technology, realizes compaction by a dynamic algorithm, which is also supposed to imitate the production stage of the material. The forces added to the particles can be manipulated, so that “sticky” particle contacts (or particle repulsion) during the production of the model material can be simulated. Also gravity effects can simply be included. This dynamic (Newtonian) simulation mechanism has no significance after completion of the simulation, hence, is not connected with the rheological properties of the cementitious model material [16]. SPACE is based on spherical particles only. However, at present a new SPACE-like system (HADES) is employed allowing the use of arbitrarily shaped particles [9] that have more serious impact on packing than so far assumed.

3.2. Hydration
The kinetic hydration model used to describe the hydration behaviour of single cement particles consist of two subsequent stages, following [17] and [18]. The first stage, in which a phase boundary mechanism controls the hydration rate, is followed by a stage in which the reaction rate is controlled by a diffusion mechanism. A uniform deposition of the gel products onto the available gel surface, a uniform decrease of the entire cement surface, and a uniform consumption of the water from the available water-air surface are assumed. Subsequently, the cement-gel interfaces as well as the gel-air and air-water interfaces remain located on concentric spheres throughout the hydration process. The simultaneous expansion of multiple neighbouring particles and the limited amount of available water will significantly affect the hydration behaviour of a single particle. For a detailed description of this process, see [19].

4. 3D PORE SIZE DISTRIBUTION – MICRO-ANALYSIS
The micro-structural analysis can be restricted to a set of parallel sections of DECs-produced model cement large enough to be representative. Three methods are available that yield 3D estimates for pore size distribution, namely application of opening operator to pore pattern (Fig. 3), of star volume to random points on pore sections (Fig. 4), and of mean free spacing to reciprocal phase of pores (Fig. 5). Hence, it is not required to use serial sectioning followed by 3D reconstruction as accomplished in [2,3,20]. Starting from a pre-defined point, Ye filled out pore space with spheres of increasing radii in the 3D reconstructed model material. This leads to biased results, however. The morphological opening operator provides a volume-based size distribution function (because of equality of areal and volume fractions.
under isotropic conditions. Caption of Fig. 6 is self-contained. Instead of Ye’s 3D pore filling approach, here the starting point is a random point system in 2D, yielding unbiased estimates.

Fig. 5 shows a simplified pattern of pores (hatched) and solid phase (white). When section image is covered by parallel line system, \( \lambda \) defines average spacing of solid clusters in 2D (average value of \( \lambda_i \)), but can be proven similar in 3D space. \( L_3 \) indicates the intercept length of the pores (indicated by dotted line on the hatched area) and when averaged defines pore size. The stereological relationship, \( \lambda = pL_3 / (1 - p) \) whereby \( p \) stands for porosity (volume fraction of pores), demonstrates \( \lambda \) to be indeed a 3D measure for pore size. See details in [21].

Fig. 3. Application of opening operator by structuring elements of increasing sizes provides a 3D volume-based size classification. Changes by openings with element of 0.3\( \mu \)m and 0.9\( \mu \)m are shown. Disappearing regions contribute to the size range involved [6].

Fig. 4. (left): Part of schematised field image in which the connected pore area (\( Y_x \)) that can be observed from random point \( x \) (light gray, at the left). Star in \( x \) measures in random directions the intercept lengths, \( l_i \) (right). Cube values of \( l_i \) are averaged to obtain the star volume \( v^* \).
5. PORE TOPOLOGY – MICRO STRUCTURE ANALYSIS

Three modern routes are mentioned here. Hilfer proposed a 3D local porosity theory (LPT) that pursues defining probability density functions for porosity and for pore connectivity in sandstone [22]. Hu & Stroeven transferred the theory to a quantitative image analysis tool with 3D stereological interpretation applicable to cement paste [23]. 2D LPT assumes in accordance with Garboczi & Bentz [24] that the 3D de-percolation threshold is at the 18% porosity level for cements with moderate fineness (Blaine 300-400 m²/kg). Fig. 6 presents data for a young cement paste. The local percolation probability denotes the probability to find a continuous path through a sample of given size. Pore space in the sample is fully connected into the Cartesian coordinate directions for $\lambda_z=1$ and is completely de-percolated for $\lambda_0=1$.

The second route is by network modelling, as used by Vogel & Roth for soils. The aim is “to keep the model as simple as possible” [3], so network models are highly idealized representations of the complex geometry and topology of pores. It is made of cylindrical pore tubes connected in a 3D face-centred cubic grid with 12 tubes connected in a node, of which part will be made redundant. Pore size distribution and topological characteristics derived

Fig. 5. Superimposed line on pores (hatched) in a section indicates 2D pore spacing $\lambda_i$.

Fig. 6. Local porosity distribution $\mu$ and local percolation probabilities $\lambda_\alpha$ for cement paste with w/c=0.4 at 3 days hydration, at the characteristic length scale of field $L=40 \mu$m [5]; $\lambda^*$ represents the local percolation probabilities in x, y and z direction ($\lambda_x=\lambda_y=\lambda_z$).
from 3D reconstruction of the material are employed for calculating the details of this network structure. Network modelling has been demonstrated an efficient tool for investigating effects of technological parameters on hydraulic behaviour of porous media [25]. Vogel & Roth [2] successfully applied network modelling to clay soils and predicted hydraulic conductivity and water retention characteristics of the soils. Comparison of simulation outcomes with experimental results revealed good agreement, indicating the hydraulic properties to be mainly governed by pore size distribution and topology. Hence, a simple network model can be generated and adapted to a predefined pore-size distribution and connectivity function.

The former approaches are of 3D nature, however can be made relying on section images only of actual cement and concrete specimens, as we have outlined for 2D LPT. We have demonstrated that de-percolation phenomenon and the relevant threshold porosity are associated with the morphological evolution of the pore space that can be characterised by mean free spacing. The mean free spacing will reduce significantly during the first period of hydration, and then gradually reaches a stable and low value at the de-percolation threshold porosity [21]. Hence, this spacing parameter gives insight into pore connectivity. Figs. 2 and 7 reveal that the bulk cement paste in the matured concrete is disconnected, while in the immediate vicinity (up to 1.5 µm from the aggregate surface) the pore space is still partly connected. Chen [4] calculated the pore connectivity in a straightforward way (serial sectioning and 3D reconstruction of DECS-produced model cement paste), according to the conventional definition of connectivity as connected fraction of porosity, for a model concrete with similar technical parameters (w/c=0.3, Blaine fineness of 300 m²/kg) at the ultimate degree of hydration. The results obtained on the basis of mean free spacing are in good agreement with the direct calculation of connected porosity, confirming that the spacing parameter reflects the geometrical aspects of the hydration process. Note that DECS-produced cement pastes (as in our work) are mostly based on a particle size range that will be roughly 50% of that in real cements, so this should be kept in mind when comparing data.

![Fig. 7. Mean free spacing plotted again distance to the aggregate interface in fresh (left) and 100 hours matured (right) conditions (w/c=0.3, specific surface area 340 m²/kg)](image)
6. PORE TOPOLOGY – MESO-STRUCTURE ANALYSIS

In this study a cherry-pit model is proposed to reflect the role that the skeleton of aggregate grains plays on concrete permeability, i.e. on pore connectivity. The aggregate grains are considered as the hard cherry pit with extremely low permeability. A shell containing the highly connected porosity is assigned to each aggregate, as part of the ITZ bordering the aggregate surface [26]. Fig. 8 shows a two-dimensional schematic of a dense random packing structure of ellipsoid aggregate grains simulated by HADES toolbox [9]. Fluids penetrate very slowly through connected pores in concrete. The total porosities constitute continuous or discontinuous paths for transport of the substances. Total amount of fluid flowing per unit of time through concrete is (at least) a function of pore size distribution, length of pore channels, and porosity connectivity. Tortuosity (length divided by projected length) of the pore channels is governed predominantly by volume fraction of aggregate grains and does not depend on the sieve curve of the aggregate grains [27]. A reduction factor has to be taken in account, which is associated with the ink-bottle effect resulting from relatively narrow passages. The ink-bottles presumably form at the transfer sections (“nodes”) between connected pores in neighbouring ITZs. Due to the patchy nature of material structure, connection in the inner layer of the ITZ will be incomplete [14].

Supposedly, the drive for simplification of models could lead to a model (again) of parallel cylindrical tubes confirming to the measured pore size distribution, and all with the same tortuosity. The tubes are narrowed on (near) contact points of aggregate grains. Numbers can be derived from CPM. Ink-bottle reduction could be assumed proportional to the amount of overlap of thin shells in CPM. This is a topic for further investigations.

Fig. 8. 2D schematic of cherry-pit model (CPM) of ellipsoid hard-core aggregate grains (left), and overlapping thin shells around each grain with connected porosity (right)

7. PRELIMINARY HADES EXPERIMENTS

SPACE is impulse-based, whereas HADES is a force-based particle packing system. Moreover, HADES allows incorporation of non-spherical particles. This is important for the simulation of crushed rock aggregate. After assessment of grain shape, properties are assigned to each particle, e.g., mass, centre of mass, moment of inertia.

As an example, crushed coarse rock aggregate is simulated by 216 irregular angular shape based on octahedrons in 4–10 mm size range. The simulated physical packing density is 0.353. A guard zone is used in the densification process for preventing physical penetration of
particles. So, effective packing density is higher. Results are presented in Fig. 9. Surface area distribution can be assigned important role in pore characterization, since percolated porosity is situated very near the surface of the aggregate.

![Image of crushed aggregate packing based on irregular octahedrons](image1)

**Fig. 9.** (left) Packing of crushed aggregate based on irregular octahedrons, and (right) its volume distribution ($V_v$).

Fig. 10(a) shows the distribution of surface area of the coarse aggregate, revealing wall effect and fluctuations in bulk value. Of course, more particles with wide range of sizes would be required for explicit assessment via cherry-pit model of proper transport properties. As a second example, Fig. 10(b) shows a compacted series of spherical and non-spherical aggregate particles simulated by HADES in the 1~20 mm size range. Spheres have been used for particles <5 mm. Some angular shapes have been used for the coarse crushed rock (5~20 mm). Four rigid boundaries and two period boundaries have been used during packing. This may demonstrate a new way in more accurately simulating particulate structure of concrete. The methodology to study hydraulic properties will be as sketched before.

![Image of simulated concrete surface area distribution](image2)

**Fig. 10.** (a) Surface area ($S$) distribution of simulated concrete of Fig. 13; (b) second example of aggregate packing by HADES
8. CONCLUSIONS

A complete and operational system for structure analysis is discussed. The developed methodology is applicable to concrete for pore structure analysis by means of pore network modelling. Pore network modelling methodology applied to SPACE- or HADES-generated concrete/cement would allow for linking up the continuously evolving material microstructure with durability issues. All measurements can rely on 2D observations. The disputable elements in the network are the “nodes”. They form the incomplete connections between the percolated pores close to the aggregate grain surfaces. Node numbers and geometrical characteristics can be studied in the jammed state of the aggregate on meso-level by employing a cherry-pit model. This is an intriguing and promising multi-scale modelling approach to study durability performance of concrete.

REFERENCES


