II International Conference on Particle-based Methods – Fundamentals and Applications
PARTICLES 2011
E. Oñate and D.R.J. Owen (Eds)

DISCRETE ELEMENT MODELING APPROACH TO POROSIMETRY FOR DURABILITY RISK ESTIMATION OF CONCRETE

PIET STROEVEN, NGHI L.B. LE, MARTIJN STROEVEN, LAMBERTUS J. SLUYS

Delft University of Technology, Delft, the Netherlands

Keywords: Concrete, DEM, pore connectivity, pore size distribution, porosimetry

Abstract. The paper introduces a novel approach to porosimetry in virtual concrete, denoted as random node structuring (RNS). The fresh state of this particulate material is produced by the DEM system HADES. Hydration simulation is a hybrid approach making use of well-known discretization and vector methods. Particle interferences (overlap) in the dynamic packing simulation and in the hydrating system are discussed. Assessment of pore characteristics like porosity, pore size distribution and pore connectivity are described. Particularly, the last descriptor, so crucial for durability estimation, cannot be determined explicitly in experiments.

1 INTRODUCTION

Concrete is a particulate material on different levels of its microstructure. Aggregate as a major constituent of concrete is generally packed into the dense random state, taking up about three-quarter of its volume. Various types of aggregate pack slightly differently; irregular shape and rough surface texture of crushed rock hamper particles arrive at similar maximum density as smooth-shaped and smooth-surfaced particles of fluvial origin do. This has obvious impact on mechanical properties of the composite. On micro-level, the binder particles in the fresh state of the material are packed in the watery environment to a density depending on the water to cement ratio (w/c). For the low and very low w/c ranges typical for high performance concrete (HPC), and for super-HPC, respectively, the binder particles are under the compaction by vibration regime also quite densely packed.

Volume density, shape and surface texture of the binder particles will influence the rheological properties in the fresh state. In engineering terms this is expressed by workability, compactability, or consistence, as it is presently denoted in the modern European standard for concrete. Consistence is nowadays controlled by adding chemical admixtures, however. Hence, in the fresh state the spatial dispersion of the aggregate as well as of the binder particles is the result of the geometric particle characteristics, the technological parameters (including mixture design) and the compaction by vibration regime. The dispersion characteristics are maintained during maturation of the concrete.

Dispersion characteristics of the aggregate control the size distribution of the binder pockets between the aggregate grains. Research has shown the surface-to-surface spacing with the closest neighbor to fall in a 1 and 200 μm range with an average of 50 μm, about the thickness of the Interfacial Transition Zone (ITZ). So, this corresponds to the (linear) size of the binder volume hydrating between the aggregate surfaces. During maturation, more and more pores de-percolate. This process depends on the dispersion of the cement particles,

which will be between randomness and order. Where binder particles form denser patches in the fresh state, pore de-percolation will occur in an early state of maturation, whereas the more dilute binder patches will lead to pore de-percolation in an advanced state of maturation or will not de-percolated at all. Research to get insight into this phenomenon would be very complicated, extremely time-consuming and thus expensive. Instead, discrete element modeling (DEM) has demonstrated to offer better perspectives. This is the type of approach followed in the research presented in this paper. For another, robotics-inspired novel approach to porosimetry in DEM-produced concrete, see [1].

2 METHODOLOGY

DEM renders possible realistically simulating binder particle dispersion in a container that represents the binder pocket between aggregate grains. The most frequently occurring situation consists of two neighbor aggregate grains with other aggregate grains on somewhat more remote locations. Hence, the prismatic container employed in DEM is traditionally provided with two rigid surfaces facing each other and four periodic surfaces. Interfacial transition zones (ITZs) will form at the rigid surfaces. For comparison purposes, a container with only periodic surfaces can be used for simulating maturing paste in bulk. Quantitatively evaluating features of gradient structures is obviously more complicated because sampling is more complex. In experimental set ups, the researcher should delineate very narrow zones on constant distances from the aggregate surface. The latter is already very complicated because the aggregate grain surface is never cut over the surface normal. This leads to biases in gradient structure observations. Moreover, to acquire enough sampling area along the aforementioned strips for reducing scatter to acceptable proportions in observations would be a very laborious job. Such sampling problems are readily solved in a DEM approach.

The assessment of the properties of the pore system in (partly) matured virtual cement paste offers a complex problem. Of the relevant parameters, total porosity is the most obvious one. Further, pore size distribution is commonly determined. However, with the eye on durability aspects, only the percolated fraction of porosity should be of interest. So, how can de-percolated pores are separated from pores that connect external surfaces of the paste specimen? For this problem, Navi [2] used the discretization approach in which the specimen is subdivided into a 3D regular lattice of cubic volume-pixels (voxels). Each voxel represents either a solid or a pore part. Then, continuity of pores between opposite sides of the specimen is determined by a process of clustering the voxels. Another approach, popular in various branches of science (biology, medicine), involves slicing the specimen and checking continuity of the pores. This serial sectioning and 3D reconstruction method was used by Ye [3] on cement paste simulated by random sequential particle addition (RSA), so with biased particle dispersion. Chen [4] improved the approach by using the DEM system SPACE. Of course, results obtained by the aforementioned methods depend on the voxel size and the slice thickness. Moreover, detection of small pores will require a large number of voxels or slices.

In this paper, a modern approach is presented for assessment of pore characteristics in virtual cement paste. The approach is based on isotropic uniformly at random (IUR) distributed nodes in pore space, and is therefore denoted by ‘random node structuring’ (RNS). An advantage over the aforementioned methods is that results can be obtained with acceptable precision without the use of a large number of nodes. Also, RNS is implemented in the DEM

system HADES, so that arbitrary shaped aggregate grains can be considered (e.g., aggregate of crushed rock or of fluvial origin). In accordance with experimental findings of Bullard and Garboczi cement particles can be of non-spherical shape. Moreover, He [5] has also demonstrated that correct ratios of surface to volume are attainable in cases of ellipsoidal particles as well as of a mixture of different polyhedron types. Generally, shape has influence on packing density of the aggregate, and thus on dispersion and spacing of the grains. This ultimately influences size of the cement pockets and degree of ITZ overlap.

So, this paper concentrates on porosimetry by RNS in virtual cement paste. The random nodes are also employed for assessment of pore size distribution ($p_o$sd) by star volume measurements [6]. The isotropic random pikes of the stars centered at the nodes measure distances to pore surface. They form the basis for unbiased local volume estimation. A volume-based $p_o$sd is directly obtained from all measurements. Of relevance is that star volume measurements can also be applied to a set of 2D random sections.

2.1 Simulation of fresh cement particles by HADES

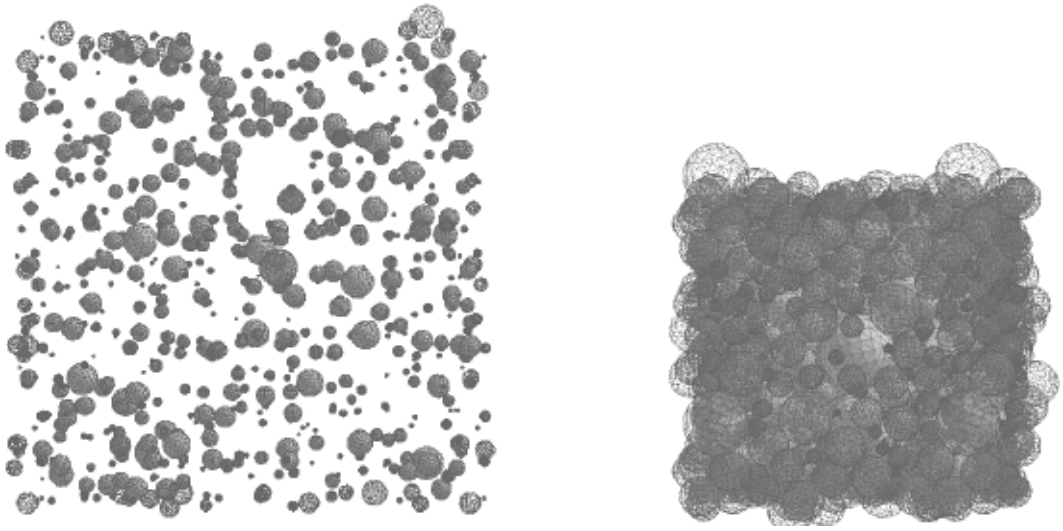


Figure 1: **S**pherical particles dynamically compacted from loose (left) into dense random state (right)

To obtain matured virtual cement paste, firstly, fresh cement particles need to be generated. In this research packing of fresh cement particles is simulated by HADES (HAbanera's Discrete Element Simulator). HADES is an advanced system for making realistic particle packing simulations, also incorporating arbitrarily grains shape. HADES is a dynamic force-based system, allowing for simulating particle packing under the influence of external forces. Mechanical interaction in HADES is based on a contact mechanism algorithm that evaluates the interaction forces exerted between segments of tessellated surfaces of neighboring particles. The contact forces are functions of distances and of areas of the segments. Several forces can be applied in this way on a particle such as spring force, cohesion force, damping force and friction force. HADES renders possible implementing particle packing in containers with periodic boundaries, simulating an infinite space, with rigid boundaries, simulating aggregate's surfaces, or with mixed conditions. Gradual reduction of container size while

particles move makes it possible achieving higher packing densities as met in practice. This is illustrated in Figure 1.

2.2 Simulation of hydration process

Several computer-based approaches for simulating the hydration process of cementitious materials have been developed the past two decades. Bishnoi [7] classified these approaches into two categories. The first, so called 'discretization approach' was developed by Bentz [8-9]. The container content is subdivided into a 3D regular lattice of cubic volume-pixels (voxels), which were randomly and in proper proportion attributed to phases. A group of "similar" voxels represents a phase in a multiphase material with eventually non-spherical particles. Results are resolution-dependent. So, large numbers of voxels are required to properly represent the microstructure, appealing to computational capabilities and time investments. Also, the approach does not consider the different kinetics in the hydration process.

The second, so called 'vector approach' does not require a sub-division of the microstructure. The method was first used by Jennings and Johnson [10]. Hymostruc3D [11] and the Integrated Particle Kinetics Model (IPKM) [2] make use of the vector approach. Calculation of particle overlap is the most laborious stage in the vector approach. Moreover, the hydration-induced growth of the cement particles also involves complicated particle interferences. The two approaches have been combined therefore in a 'hybrid' system.

Initially, hydration simulation is based on the IPKM. This holds for the tri-calcium silicate (C_3S) in the fresh cement. For that purpose, the C_3S is modeled as spherical cores. As a result of the reaction with water, calcium silicate hydrate (CSH) and calcium hydroxide (CH) is formed. The CSH amount formed during a short time lap is modeled as a product layer surrounding the C_3S particle's core. Part of the CSH amount ('in' product) replaces the C_3S amount that has reacted and the remaining CHS amount ('out' product) precipitates on the particle's surface. The CH is assumed to either diffuse and nucleate randomly in pore space or precipitate on the surface of the existing CH grains. CH is also modeled as spherical particles. The models of different products of cement particles in the IPKM are illustrated in Figure 2.

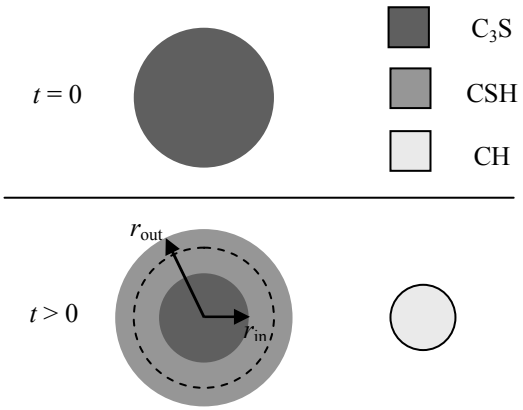


Figure 2: Models of different products of cement particles in the IPKM

Next, some improvements on the effects exerted by particle interferences (overlap!) are implemented by means of the discretization technique. This leads to a re-calculation of the

thickness of the new product layer on the declining free surface of the particles. The latter is determined by means of the sampling point method [12]; each particle's surface is sampled by a number of uniformly at random distributed points [13]. The free surface fraction equals the ratio of the number of sampling points not interfering with other particles over the total number of sampling points. For an efficient checking procedure for such interferences, neighborhood should be defined. This is accomplished with the 'cell method' [7] (or grid subdivision method) in which the simulated space is subdivided into small cubes named 'cell'. Each cell contains a list of the particles that interfere with the cell. Therefore, to determine which particles have to be taken into account in interference situations, only the cell as the neighborhood should be investigated. Figure 3 illustrates this for the 2D case.

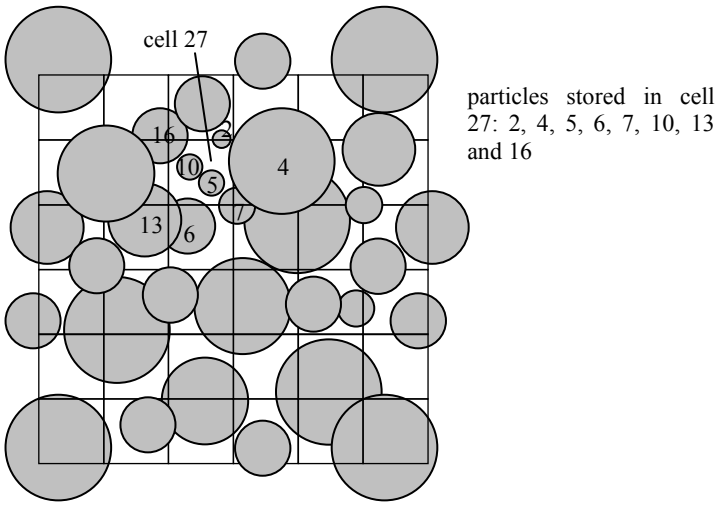


Figure 3: Particle interference check by the cell method (2D)

For calculating the thickness of the new product layer precipitating on hydrating particles in complicated particle interference situations, an efficient numerical procedure is proposed. In it, the simulated space is sub-divided into very small cubes called 'voxels'. Each voxel represents either one of the two possible phases, *i.e.,* solid and pore. Each voxel's phase is updated at each time step of the hydration process. With each trial value of the layer thickness in the iterative procedure, the number of voxels that switch from 'pore phase' into 'solid phase' due to the formation of the new layer is determined. The iterative procedure will stop when the total volume of the aforementioned switching voxels corresponds to that of the diffused product.

3 RANDOM NODE STRUCTURING (RNS) METHOD

3.1 Algorithm

RNS is a general approach, so it can be used for virtual cement paste simulated by the vector approach, the discretization approach or the aforementioned hybrid system. It starts by generating IUR dispersed nodes in container space by a pseudo-random generator algorithm. Then, the nodes situated in solid phase are eliminated from further consideration. This yields IUR dispersed nodes inside pore space as a detection system of the 3D capillary pore system

in cement paste. The next step is a structuring process for the node system in which the relationships among the nodes are built up by unobstructed straight line segments between pairs of nodes. Figure 3 presents a 2-D scheme for RNS.

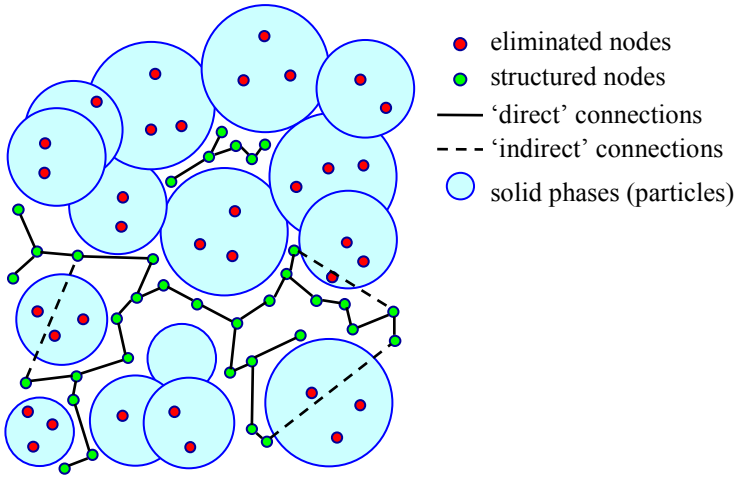


Figure 4: Structure of random nodes in RNS method

When two nodes can be connected by a straight line that does not intersect with any part of the solid phases, such two nodes have 'direct connection'. This initiates a node-clustering process. A cluster involves a group of nodes each of which is directly connected to at least one of the other nodes. When two of such clusters during this process can be directly connected between two nodes from the respective clusters, they merge into a new one. Hence, once the clustering process is finished, there are a number of structured clusters of nodes. In each of these clusters, the nodes are mutually connected, which represents the connectivity among pores in the virtual cement paste. Based on the node *structure*, pore characteristics such as porosity, degree of percolation, pore location distribution and pore size distribution are assessed.

3.2 Implementation

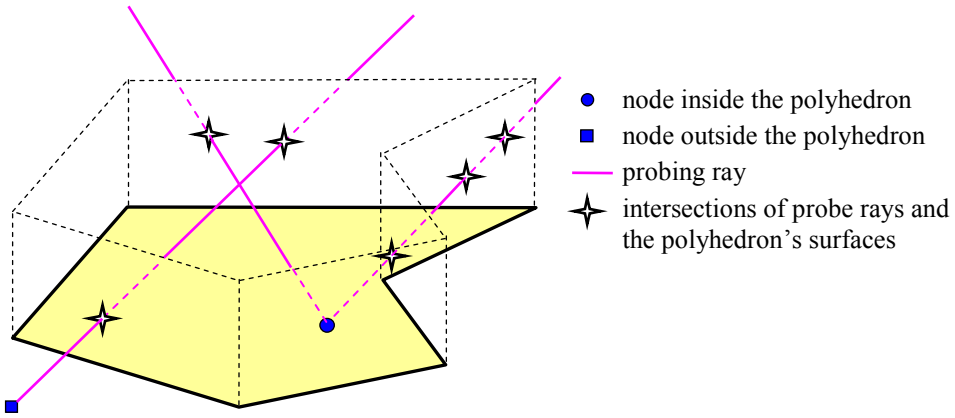


Figure 5: Determination whether a point is inside or outside a polyhedron

Elimination of nodes in solid phases. As mentioned above, to obtain nodes IUR distributed in pore space, an elimination process is required of the nodes interfering with solid phase. For spherical cement particles this can easily be accomplished. For voxel-based cement particles this is also easily carried out by determining the phase of the voxel in which the considered point is situated. Figure 5 illustrates the case of a cement particle simulated by a polyhedron. A probing ray emanating from the considered node in a random direction is generated. In fact, this ray is a line segment (limited by two ends) and its length must be long enough to possibly go through the polygon. If the number of intersections of the ray and the polyhedron's surfaces (polygons) is odd then the point is inside the polyhedron, otherwise it is outside the polyhedron.

Detection of connection between two nodes. As mentioned above, the RNS method involves an IUR system of nodes that are pair wise connected by unobstructed line segments. The line segment will be considered as being unobstructed if it does not have any intersection with neighboring particles. In the case that solid phases are spherical, the intersection detection reduces to that of a line and a sphere. In the case that the discretization approach is used for simulating the microstructure, a series of points with constant spacing is distributed along the line segment. The segment line will be unobstructed if there is no point situated in any voxel representing a solid phase. When particles have polyhedral shape, the detection procedure is the same as earlier described. Since two ends of the line segments are outside particles, the number of intersections is only either zero or an even number. If the number is zero then there is no intersection. Otherwise, such an intersection exists.

Improvements in computation with localized and parallel computing processing. The RNS method centers on finding direct connections between neighboring nodes. The cell method presented in part 2.1 is utilized for this purpose. Each cell contains a list of the particles that interfere with the cell and a list of associated nodes. The cells render possible executing localized operations to improve the speed of computation. So, the clustering process is first applied locally in each cell. Thereupon, the clustering process is applied in the whole simulated space. Moreover, since local clustering processes can be implemented independently and simultaneously, 'parallel computing processing', the advanced feature of some programming languages, is also applied to speed up the computations.

Detection of pore connectivity. Considering a cubic sample pocket of cement paste, two types of capillary pores can be distinguished. The percolated pores form continuous entities connecting two opposite sides of the specimen. The de-percolated pores or the isolated pores might be connected together but are not connected to both sides of the pocket. The connectivity of pores is defined as the volume ratio of percolated pores to total of capillary pores. To complete detection of pore connectivity, also nodes should be uniformly at random distributed on the opposite outer surfaces of the specimen and incorporated in the clustering process. If there is a cluster of nodes containing at least one node in both end surfaces, then this cluster represents a percolated pore channel. In analyzing characteristics of the pore system, e.g., porosity, connectivity, pore size distribution, the nodes situated in the two end surfaces are not taken into account, however.

4 MEASURING PORE SIZE

In 2D sections of real or virtual cement paste alike, the most direct way of obtaining 3D local volume information on irregularly shaped pores of is by way of the mathematical morphology operator 'opening' [14]. This has been demonstrated in [15-17]. The underlying requirement of structural isotropy can be expected to be violated however inside the ITZ, so the method should be applied to bulk cement only. A next option is making star volume measurements, a method widely employed in life sciences [6]. This will be discussed later.

With computer-based models to simulate the microstructure of cement paste, a 3D pore structure can be produced and visualized by either one of the earlier approaches. Hence, a direct 3D assessment method for pore size would be attractive. Ye [3,18] filled up the reconstructed pores by spheres of successively increasing size, starting from a pre-determined point. This has been indicated leading to biased results [16,19]. The earlier mentioned technique of star volume measurements can be applied in 3D, however. Applying the RNS method, we can employ the random nodes as the centers of 3D 'stars' in the star volume method. Then, a large number of 'pikes' extend from the star center in isotropic at random directions to reach the nearest pore surfaces. Each of their lengths, l_i, is measured, whereupon local pore volume, V_j, is obtained from $V_j = 4\pi\overline{l_i^3}/3$ ($\overline{l_i^3}$ stands for average of values of l_i^3), so that local pore size (i.e., diameter) equals $2\sqrt[3]{\overline{l_i^3}}$ [6]. Next, the volume-based cumulative pore size distribution function is constructed from all local pore size measurements, whereupon the volume-based pore size distribution function, $p_o$sd, is obtained through differentiation of the cumulative function. This is the most appropriate method for 3D assessment of $p_o$sd in virtual cement.

5 EXAMPLES

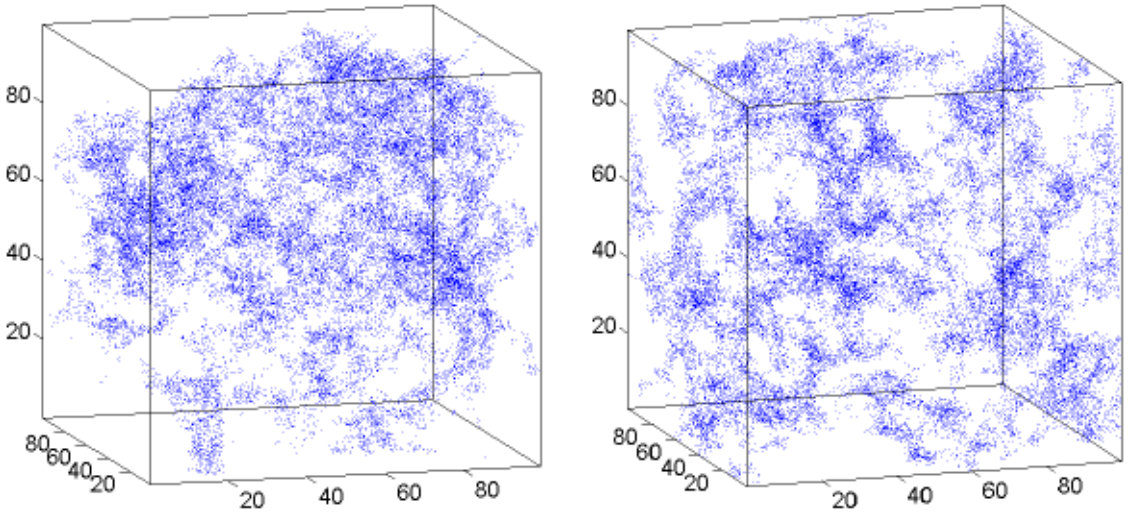
Two cubic pockets of simulated cement paste are considered in this study. The characteristics of the two pockets are shown in Table 1 (P=periodic, R=rigid boundary). Fresh cement particles were generated and packed with the HADES systems. The initial size of the pocket was eight times reduced in the dynamic packing process. Next, the hybrid hydration simulation approach described in 2.2 was implemented. Next, the pore structure of the samples is investigated by RNS at ultimate degree of hydration (DOH). Due to shortage of water for chemical reaction, the hydration process of the samples stops at ultimate DOH of 0.727.

Table 1: Characteristics of simulated cement samples

	Boundary conditions	Diameter range (μm)	Parameters of Rosin-Rammler distribution [12]		Specific surface area (cm^2/g)	Size of pocket (μm)	Intial w/c ratio	Number of particles
			b	n				
C1	6P	1~30	0.07	2.599	1838	100	0.304	4060
C2	4P + 2R							

Figure 6 presents the nodes distributed IUR in pore space of the two cement pockets. Based on these systems, porosity of the C1 and C2 pockets was found to amount 5.57 % and

5.59 %, respectively. The associated connected pore fractions of the C1 and C2 pockets were 98.53% and 90.7%. Figure 7 shows the gradient structures of distribution of pore volume as a function of distance from the rigid surface. Figure 8 presents examples of the pore size distribution assessed by the star volume method. Figure 9 is the sensitivity analysis revealing the influence of the numbers of the IUR nodes initially generated on connected fraction of porosity in the pocket C1. It can be seen that the value of connected fraction raises with the increase of the number of nodes to reach a plateau value when the number is exceeding 1 million. The number of the IUR nodes, however, does not have much influence on the porosity and the pore size distribution. For example, the porosity is about the same for $0.2x10^6$ and $2x10^6$ nodes, while the pore size distribution curves for these numbers nearly coincide as shown in Figure 10.



(a) C1 (b) C2

Figure 6: System of IUR nodes distributed in pore space of the two cement paste pockets

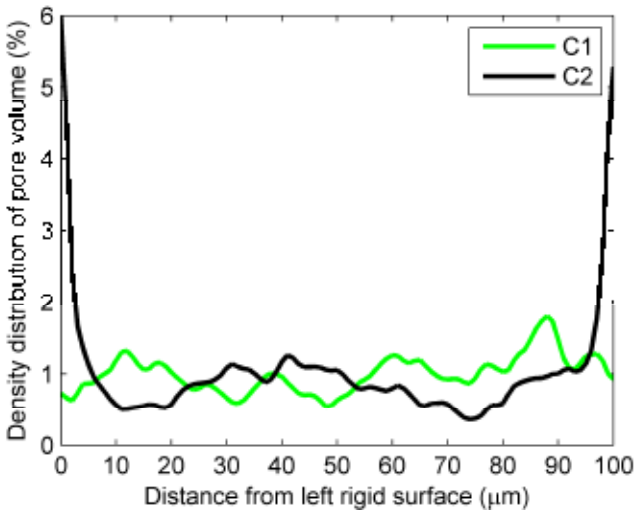


Figure 7: Density distribution of pore volume as function of distance from the left rigid surface

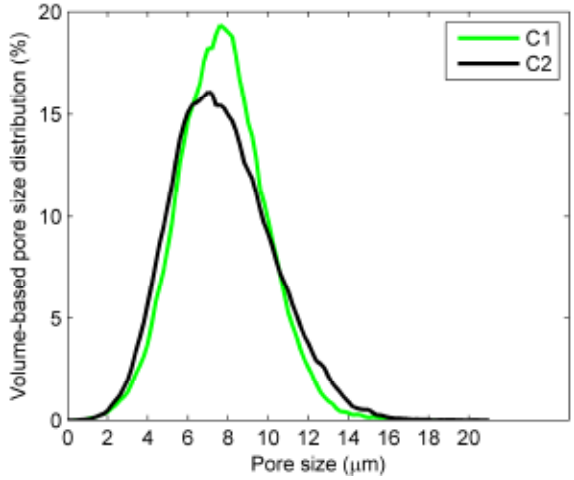


Figure 8: Volume-based pore size distribution

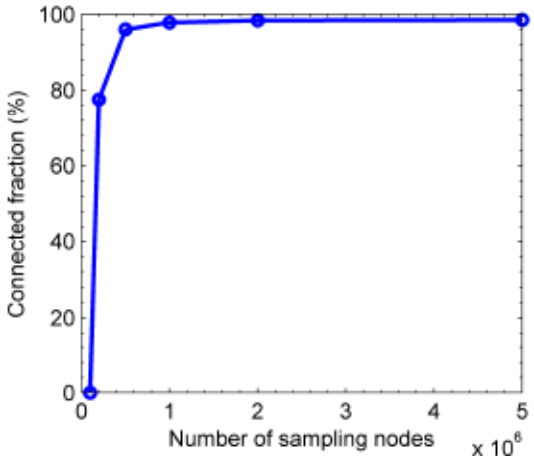


Figure 9: Sensitivity of connected fraction of porosity on number of IUR sampling nodes

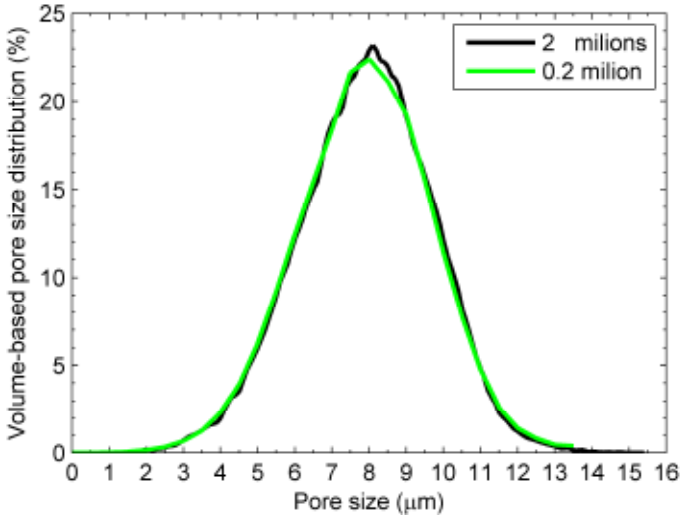


Figure 10: $p_o$sd functions for different numbers of sampling nodes

6 CONCLUSION

A dynamic DEM approach for simulation of fresh cement paste pocketed between aggregate grains in concrete is presented. The HADES system that is employed renders possible simulating artificial grain shape. A hybrid approach is presented for simulation of the hydration process, combining the well-known vector and Integrated Particle Kinetics approaches. Special procedures are incorporated to account for particle interference problems in the fresh and hardened states. The proposed RNS method involves generation of IUR dispersed nodes in pore space, whereupon a process of node clustering is implemented by finding straight line connections between nodes. The final state presents the full network structure of pores in the maturing cement paste. Additional application of star volume measurements at node locations leads to the pore size distribution, while porosity is governed by the relative number of nodes in pore space. Pore connectivity is derived from the network structure. Hence, all pore characteristics relevant for durability estimation can be obtained. An example demonstrated the operational capabilities of the RNS method. The approach is potentially very versatile, since different grain shapes can be considered and from the simulated cement pocket bulk data can be derived or the pocket can have ITZs near neighboring rigid aggregate grain surfaces. Compared to quantitative image analysis approaches to real concrete, the RNS approach combines economic and reliability qualities, the latter because of present-day advanced computer facilities. Moreover, it offers insight into pore connectivity that experiments cannot, or at least cannot in an equally economic and reliable way.

REFERENCES

[1] Stroeven P., Nghi L.B.N. Studying percolated porosity in concrete by DEM. *Proc 1st Interquadrennial ICF Conference in Middle East and Africa: Processing, Performance, and Failure Analysis of Engineering Materials*, Luxor (14-17 Nov. 2011).

[2] Navi P., Pignat C. Simulation of cement hydration and the connectivity of the capillary pore space. *Advanced Cement Based Materials* (1999) **4**(2):58-67.

[3] Ye G. *Experimental study and numerical simulation of the development of the micro-structure and permeability of cementitious materials*. PhD Thesis, Delft University of Technology (2003).

[4] Chen H.S., Stroeven P., Ye G., Stroeven M. Influence of boundary conditions on pore percolation in model cement paste. *Key Engineering Materials* (2006) **302-303**:486-492.

[5] He H. *Computational Modelling of Particle Packing in Concrete*. PhD thesis, Delft University of Technology (2010).

[6] Gundersen H.J.G., Bagger P., Bendtsen T.F., Evans S.M., Korbo L., Marcussen N., Møller A., Nielsen K., Nyengaard J.R., Pakkenberg B., SøRensen F.B., Vesterby A., West M.J. The new stereological tools: Disector, fractionator, nucleator and point sampled intercepts and their use in pathological research and diagnosis. *APMIS* (1988) **96**(7-12):857-881.

[7] Bishnoi S., Scrivener K.L. μic: A new platform for modelling the hydration of cements. *Cement and Concrete Research* (2009) **39**(4):266-274.

[8] Bentz D.P. Three-dimensional computer simulation of portland cement hydration and microstructure development. *Journal of the American Ceramic Society* (1997) **80**(1):3-21.

[9] Bentz D. Modelling cement microstructure: Pixels, particles, and property prediction. *Materials and Structures* (1999) **32**(3):187-195.

[10] Jennings H.M., Johnson S.K. Simulation of Microstructure Development During the Hydration of a Cement Compound. *Journal of the American Ceramic Society* (1986) **69**(11):790-795.

[11] van Breugel K. *Simulation of cement hydration and formation of structure in hardening cement based materials*. PhD thesis, Delft University of Technology (1991).

[12] Stroeven P., Stroeven M. Reconstructions by SPACE of the Interfacial Transition Zone. *Cement and Concrete Composites* **23**(2-3):189-200.

[13] Weisstein E.W. *Sphere Point Picking*. From MathWorld--A Wolfram Web Resource. http://mathworld.wolfram.com/SpherePointPicking.html.

[14] Serra J.P. *Image analysis and mathematical morphology*. Academic Press (1982).

[15] Hu J., Stroeven P. Depercolation threshold of porosity in model cement: approach by morphological evolution during hydration. *Cement and Concrete Composites* (2005) **27**(1):19-25.

[16] Hu J., Stroeven P. Proper characterization of pore size distribution in cementitious materials. *Key Engineering Materials* (2006) **302-303**:479-485.

[17] Stroeven P., Hu J., Koleva D.A. Concrete porosimetry: Aspects of feasibility, reliability and economy. *Cement and Concrete Composites* (2010) **32**(4):291-299.

[18] Ye G., van Breugel K., Fraaij A.L.A. Three-dimensional microstructure analysis of numerically simulated cementitious materials. *Cement and Concrete Research* (2003) **33**(2):215-222.

[19] Hu J. *Porosity in concrete – morphological study of model concrete*. PhD Thesis, Delft University of Technology (2004).