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### Porosimetry by DraMuTS applied to DEM-produced cementitious materials

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#### Abstract

Porosimetry of matured cementitious materials is of high interest, because durability is governed by the capillary pore network structure. Experimental approaches are time-consuming, laborious and thus expensive. Fast developments in computer technology make it nowadays possible quite realistically producing cementitious materials in virtual reality by DEM. This renders possible studying the materials in a reliable and economic way. This involves the production of fresh material, hydration to get the matured material, delineation of the capillary pore network system, and assessment of pore characteristics. Finally, a model can be constructed correlating the obtained pore characteristics with global hydraulic properties of the cementitious material. The paper concentrates on the porosimetry methodology, however is illustrated by some results obtained this way on (blended) cement paste.

Keywords: Concrete, porosimetry, DEM, hydration, pore topology, pore size

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# Introduction

The relevance of porosimetry of cementitious materials is obvious, because durability is governed by the capillary pore network structure in hardened concrete. Fast developments in computer technology make it nowadays possible quite realistically producing cementitious materials in virtual reality by the Discrete Element Method (DEM). DEM has been developed the past three decades in various fields, including concrete technology. A survey is given in [1]. This renders possible studying the materials in a reliable and economic way. This involves the production of the fresh material by a dynamic packing algorithm, simulated hydration to get the matured material, delineation of the capillary pore network system, and assessment of pore characteristics.

For the first operation, the dynamic concurrent algorithm-based DEM, denoted HADES, is introduced. It provides for packing arbitrarily-shaped particles from the dilute to the dense random state, whereby particle dispersion is realistically simulated. The dense random packing state for fresh concretes is relevant for aggregate grains as well as for binder particles in the lower water to cement ratio range. Popular in concrete technology are so called random sequential addition (RSA) systems; however, they ignore particle interferences so characteristic at practical particle densities in the aforementioned dense random packing state. As a result, particle dispersion in the virtual material will be biased [1, 2]. This will be detrimental for reliably predicting structure-sensitive properties of concrete. This holds for the dispersion of the aggregate grains. This influences the characteristics of the pore network. Also the process of pore de-percolation has been demonstrated structure-sensitive [3].

In the second operation, the fresh material is hydrated by the cement hydration model CemHydSim. Herein, the Integrated Particle Kinetics Model (IPKM) for simulating hydration of  $C_3S$  grains [4] is upgraded to a multi-compound hydration model. Moreover, the hydration process can also consider grains of incinerated, silica-rich fine-grained vegetable waste (in particular rice husk ash [5]). Hydrating particle's expansion in the complicated situation of interfering particles is assessed by a new numerical approach. Also, the microstructure of hydrated cementitious material is stored and visualized by a 3D voxel system (*cf.* pixels in digital image), whereby each voxel represents a particular phase.

The third operation concerns the exploration and delineation of the complete capillary pore network structure in the hardened (blended) Portland cement paste, which is either pocketed between nearby or between more remote aggregate grain surfaces. For that purpose, Double Random Multiple-Tree Structuring (DraMuTS) has been developed. Inspiration came from the rapidly-exploring random tree (RRT) approach in robotics. Path planning is implemented by generating a 'virtual tree' system that includes sets of nodes ('vertices') and lines ('edges') that connect pairs of nodes, like branches of real trees. In robotics, this tree grows incrementally and randomly in 3D. An upgraded multiple-tree generating system is developed, whereby trees can merge, when relevant. Moreover, random growth is victimized for increasing speed of simulation. Finally, all capillary pores can be explored and delineated this way.

In the last operation, the geometric and topological pore network properties are assessed. For that purpose, a second system of random points is generated by DraMuTS, of which only the points located inside the pore network structure are employed for assessment of pore characteristics. The tree characteristics allow distinguishing between continuous channels, connecting outside surfaces of the cement paste pocket, dead-end pores branching off these channels and isolated pores. The respective point fractions govern the associated pore volume fractions. Pore size distributions are directly obtained by application of star volume measurements. Pore tortuosity can be determined after applying a smoothening technique to the pore channels. Pore length and diameter may finally be incorporated in a model for predicting transport through the concrete.

# **DEM by HADES**

To produce packing of particles, the mutual interference between grains as well as between grains and the boundaries of the container should be modeled. In practice, this is achieved by two strategies, involving a static or a dynamic stage. In the first, overlap of particles is corrected by locally shifting the involved grains [6]. In a dynamic solution all particles move and rotate and upon overlap develop repulsive forces that will eliminate the overlap. Intelligent algorithms are necessary for improving the economy of the approach. The result is a so called dynamic solution for a concurrent algorithm-based simulation method. The procedure starts by dispersing the grains in a large container of which the size is gradually reduced during the dynamic stage. When the desired volume fraction is achieved, the procedure stops (Fig. 1). Alternatively, densest particle packing can be realized whereby particles are in the so called dense random packing state. This is a relevant situation for aggregate packing. Comparison between experimental results and simulation results on densest aggregate packing of differently graded mixtures yielded satisfactory agreement [6].





Figure 1: Compressing spherical particles from loose (left) to high (right) density with gradual reduction of container size (and change of scale) in case of four periodic boundaries and two rigid ones, used to simulate cement paste between two nearby aggregate surfaces.

Most of the DEM and RSA approaches only accept spherical grains. However, HADES renders possible realistically simulating particle shape; this complicates the algorithms for particle interference, of course. The grains are surrounded by a thin boundary zone. Upon starting overlap of boundary zones of nearby grains, the part of the involved surfaces of both grains is activated. The surface is tessellated for that purpose by triangles [6]. Depending on the local degree of boundary layer penetration, the resulting repulsive force will grow and will cause the particles to separate again. This strategy has been successfully applied for simulating packing of river gravel as well as of crushed rock aggregate. Of course, the grains of a specific aggregate will be represented by a single "average" shape or a mixture of a very limited number of shapes. Fig. 2 presents an example of grain packing simulation obtained by HADES [7].

Also for binder particles at low water binder ratio, volume fraction can be as high as 0.6. The procedure is the same as described for the aggregate. The particle size distribution of the

cement is generally scaled down by a factor of about  $1/3^{rd}$  to significantly improve the economy of the approach. The same approach as for aggregate is used for simulating non-spherical Portland cement grains as experimentally found in [8]. The surface (*S*) to volume (*V*) ratio is as a consequence deviating from that of a sphere and will have impact on the hydration process. Fig. 3 shows that for polyhedral packing, the same *S/V* curve could be reproduced [7]. The far more complicated hydration algorithms in particle interference situations are still in development, however.



Pentahedron

Octahedron





Figure 3: HADES simulation of 1000 packed multi-size polyhedrons in 10~50 µm size range; experimental regression results (right) and visualized structure of compacted grains (left).

## Hydration simulation

Bishnoi and Scrivener [9] distinguish the discretization approach developed by Bentz [10] from the vector approach first used by Jennings and Johnson [11]. The latter approach is also used by Navi and Pignat and denoted Integrated Particle Kinetics Model (IPKM). The first approach is resolution-dependent, the second is more time-consuming in solving the complicated particle interference situations. Presently in our studies, the first hydration part is accomplished by the IPKM approach, whereas volume-pixels (voxels) are employed in the

second stage. The method is denoted CemHydSim and is described in [12]. Recently, the method is extended to cover the two major compounds of PC, *i.e.*  $C_3S$  and  $C_2S$ . See, Fig. 4. Intelligent algorithms (*e.g.*, for neighborhood definition) are developed for reducing computer time. Also particles of a silica-rich mineral admixture (Rice Husk Ash (RHA)) are included. Finally, inert particles will be added to the system.



Figure 4: Particle models of cement, pozzolanic admixture and hydration product.

# DRaMuTS pore delineation stage

A random point system is superimposed on the virtual material. Only those inside the pores are considered. A path planning algorithm inspired by the rapidly-exploring random tree (RRT) approach is applied to the points resulting in the growth of tree-like structures consisting of the points and connecting lines. The efficiency of the RRT approach is improved at the expense of violating randomness. Moreover, multiple trees are growing simultaneously from randomly chosen or selected "seeds", whereupon connected trees will merge. In the measuring stage, another random point system is superimposed, which resulted in the name: Double Random Multiple-Tree Structuring (DRaMuTS). Details of the methodology are described in [3, 6, 13]. By increasing the number of points, the connectivity in the pore system is enhanced approaching a plateau value depending on the fineness of the capillary pore system between about  $10^4$  and  $10^5$  tree edges. Since the finest pores probably do not contribute significantly to the transport system, a lower sensitivity level could be selected for practical purposes making the approach even more economic. Fig. 5 shows tree structures for PC and gap-graded blended PC for all detected pores (left) and only for the main trunks (right: directly connecting outside surfaces of the specimen).

# DRaMuTS pore measuring stage

The second stage starts by the aforementioned generation of the second random point system of which the ones outside pore space are removed. The point fraction associated with the channels equals its volume fraction or continuous pore fraction. Hence, volume fraction of pores branching off the main channels and volume fraction of the isolated pores are similarly determined. With a developed algorithm that finds all ends of pores, the full pore topology is assessed. The effect of the aggregate grain surface on the distribution of total pore volume can thus easily be displayed. This gradient structure can be studied as function of interesting technological parameters. As an example, the effect of gap-grading by a

mineral admixture (*i.e.*, RHA) has been studied. It could be demonstrated this way that the pore peak value zone inside the ITZ was narrowed due to blending, as Fig. 6 convincingly demonstrates.

Next, the random points are provided with a star, *i.e.* from each point lines run in random or systematic directions to the nearest surface of the pore, forming the stars' pikes. Length  $l_i$  of all pikes are measured per point, cubed and averaged. Twice the third root value is an

estimate of the local pore diameter,  $d_i$ . Hence,  $d_i = 2\sqrt[3]{l_i^3}$ . The method is called star volume measuring and is developed basically for estimating pore size from 2D sections [14]. However, in the 3D reality of the virtual material it can be directly applied. The collected volumes of all such representative spheres can be used for the construction of a volume-based pore size distribution function (PoSD), which renders possible studying the effects of technological measures on the PoSD. As an example, the favorable effect of blending by a gap-graded mineral admixture (*i.e.*, RHA) on the PoSD has been revealed this way [5, 6] (Fig. 7).



Figure 5: Pore delineation in 100µm cubes for PC (top) and gap-graded blended-PC (bottom). All capillary pores are shown (left) as well as the continuous trunks only (right).

With the eye on possible application of pore size measurements in a transport-based model, it seems more logic determining the local pore throat. This is defined as the smallest pore area of random plain intersections through a random point. In such a random section a 2D star is defined by a random or systematic set of pikes in the section plane. Pike length  $l_i$  is determined per point, squared and averaged. The area of the cross section is given by  $\pi l_i^2$ . The area-based pore throat size distribution function is obtained by collecting all local pore throat area measurements. Curves are not fundamentally different from PoSD.

A final operation could be smoothening of the zigzag lines in channels to get a more appropriate measure for their tortuosity. This is being executed at the moment. Relevance is derived from the expectation that pore size (throat distribution) and pore tortuosity (real length divided by projected length) would be the major parameters in a transport model.



Figure 6: Gradient structures of total porosity as influenced by gap-graded blending by RHA. Pc=Portland cement, Po20 is Portland cement with 20% RHA, W40 = *W/B* or *W/C*=0.40).



Figure 7: Pores are coarser inside the ITZ zone in PC, however disproportionately refined by gap-graded blending (MZ is middle zone, between ITZs, Pc=Portland cement, Po20 is Portland cement with 20% RHA, W40 = *W/B* or *W/C*=0.40).

#### Conclusions

The completed methodology of HADES-CemHydSim-DRaMuTS would render possible elaborating in an economic way comprehensive studies on influences of technological parameters on geometrical and topological pore characteristics. HADES simulation would make the outcomes reliable and superior in comparison to similar information obtained from in

concrete technology quite common RSA systems. The favorable strength experiences with gap-graded blending by RHA in real concrete [15] are supplemented in the present virtual experiments as to porosity. This will exert also positive effects at to durability.

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