ANALOGUE SIMULATION BY DEM OF MATERIAL STRUCTURE FOR PROPERTY ESTIMATION OF CEMENTITIOUS MATERIALS

Piet STROEVEN¹, Huan HE² and Nghi L.B. LE¹
¹Faculty of Civil Engineering and Geosciences, Delft University of Technology, Stevinweg 1, PO Box 5048, 2600 GA Delft, the Netherlands, e-mail: P.Stroeven@tudelft.nl
²GeMMe, Minerals Engineering-Materials-Environment, University of Liège, Chemin des Chevreuils 1, 4000 Liège, Belgium.

ABSTRACT
Realistic simulation of particulate materials like concrete on meso- as well as micro-level is nowadays possible by fast developments in computer technology. This would be a more economic way than by physical experiments, which are more time-consuming, laborious and thus expensive. This concern the production of the aggregate structure or of the fresh binder material pocketed between aggregate grains. In the latter case, it should be followed by hydration to get the matured material. A subject of major relevance is porosimetry. This requires techniques of delineating the capillary pore network structure for the assessment of topological and geometric properties. By combining such features with hydraulic properties, a model could be designed for estimating transport properties of concrete. Influences of technological parameters on packing characteristics are of interest for optimum packing, strength and durability of cementitious composites. The paper will concentrate on packing problems and the resulting pore network structure. Some results will be presented for illustrative purposes.

Keywords
Concrete, aggregate, particle packing, DEM, hydration, pore network.

INTRODUCTION
Packing capacity of aggregate in concrete is obviously of engineering and of economic interests. Although ample research data are available, developments in concrete technology - particularly in the high performance range - ask for additional research. This can now easier, but still reliably and in a more economical way be performed by the discrete element method (DEM). The versatile HADES system is used for that purpose. This is a dynamic concurrent algorithm-based system [1,2]. It renders possible considering arbitrarily shaped packed particles from the dilute to the dense random state [2]. The particle packing problem is among the oldest topics in mathematics and physics. So, additional information is available, although mostly dealing with spheres. More recently, also DEM has been employed for packing of ellipses in 2D [3] and ellipsoids in 3D [4]. The fundamental difference with concrete technology is the matrix. In physics, the densest packing is pursued. In concrete technology, however, the gravel grains are embedded in a cementitious matrix with a certain consistence, which upon hydration binds the aggregate grains together. Packing density is thus inevitably lower than found in such physics studies. Coordination number will be reduced as well. Nevertheless, we are interested to see whether particle shape will exert similar effects on packing density and coordination number as found in physics.
Studies of cement packing in the fresh state are performed in a similar way. The loose random packing density is about the maximum level required for such binder packing. Hydration algorithms are required for studies of hardened microstructures and porosimetry. We have covered spherical particles as well as ellipsoidal ones with various aspect ratios and polyhedral grains of different types. The two categories could represent aggregate of fluvial origin or of crushed rock, respectively. Moreover, they could represent cement particles, which were experimentally revealed to be non-spherical [5]. 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 [6] 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). Hydrating particle’s expansion in the complicated situation of interfering grains 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.

GENERATION OF PARTICULATE STRUCTURE

The HADES system is a dynamic concurrent algorithm-based DEM, in which particle interference is an integral part. In a dynamic system, this is accomplished by having the particles “moving around”. The procedure is such that the grain mixture in which the particles have a desired shape is dilute dispersed in a container. Hence, in this stage a random sequential addition (RSA) algorithm can be of profitable use. For generation of the densely packed aggregate structure, this approach yields a biased solution, despite being quite popular in concrete technology [7]. Next, grains are provided with a thin guard zone and are set to move according to a Newtonian system for linear and rotational motions. Surfaces of the grains are tessellated into a triangular system. Upon overlap of guard zones of colliding grains, interaction forces between the associated tessellated surface elements develop. This force is a function of penetration distances and areas of the segments and is integrated for the activated part of the surface. Such forces can be spring-like (leading to repulsion), cohesive (leading to attraction) or representations of damping (due to energy losses in the system) and friction effects.

The grains also similarly interact with the mould of the container, whereby the container boundaries can be periodic, rigid, or a combination. During this dynamic stage, the container shrinks in size until the desired volume fraction of the grains is attained. The guard zones lead to small boundary gaps between the grains (Fig. 1). This has been at least partly compensated by a growth process of the grains. Maximum packing density could have been obtained by application of randomized shifts and rotations in places where overlap is found such as

Fig. 1. Growth for eliminating gap by guard zone in HADES simulation.
Fig. 2. Packed structures of arbitrary octahedra (maximum size about 5 mm) with rough surface area texture at different growth levels of 0.1, 0.2, 0.3 and 0.4 mm, respectively.

performed in a static DEM. However, in concrete technology this would be an irrelevant action, because of the presence of the cement matrix between the aggregate, while the highest density in the binder would be that of the loose random packing state. An example of loose random packed crushed rock aggregate structure represented by polyhedra subjected to growth is shown in Fig. 2.

In physics experiments similar structure generation procedures pertain. In [3], ellipses are generated by RSA procedures, whereby the perimeter is discretized (leading to polygons). Thereupon, growth is step-wise applied. Next, they locally shift and rotate the polygons in the static DEM approach for elimination of overlap. These growth steps are applied until total overlap is exceeding a pre-fixed level. [3]. In our research we also stop when overlap is exceeding a certain limit.

HYDRATION GENERATION

Bishnoi and Scrivener [8] distinguish the discretization approach developed by Bentz [9] from the vector approach first used by Jennings and Johnson [10]. The latter approach is also used by Navi and Pignat and denoted Integrated Particle Kinetics Model (IPKM) [6]. The first approach is resolution-dependent and 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 to represent the microstructure. The method is denoted CemHydSim and is described in [1]. Recently, the method is extended to cover the two major compounds of Portland cement (PC), i.e. C₃S and C₂S. See, Fig. 3. Intelligent algorithms (e.g., for neighborhood definition) are employed 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.

SHAPE REPRESENTATION

The economy of the experiment requires selecting a “representative” shape for a type of aggregate particles, as shown in Fig. 4. For aggregate of fluvial origin, ellipsoidal grains are selected, whereas for crushed rock aggregate, polyhedra are taken. In both cases, more than one type of grain shape can be selected, of course. For cement grains, similar shape representation is accomplished, so that surface area to volume of the grains is in agreement with experimental findings [5]. This is shown for polyhedra in Fig. 5 [11]. For ellipsoids, see [2].
Fig. 3. Particle models of cement, pozzolanic admixture and hydration product.

Fig. 4. Representation of river gravel and crushed rock aggregates by ellipsoids and polyhedra, respectively [2].

Fig. 5. HADES simulation of 1000 packed multi-size polyhedra in 10–50 µm size range; experimental regression results (right) and visualized structure of compacted grains (left).
EFFECT OF GRAIN SHAPE ON PACKING

In the 2D physics experiments on multi-size ellipses in the “jammed state”, authors in [3] found a volume density of 0.84 for circles with a polydisperse distribution. At declining value of the aspect ratio $\lambda$, the density increased to about 0.9 for $\lambda \sim 0.7$, whereupon it declined again. For $\lambda < 0.25$, the packing density was even below that of the circles. At the proper sensitivity level for the assessment of contacts between ellipses, the average contact number increased from 4 for circles to a plateau value of 5.7 that was reached at $\lambda \sim 0.6$ (6 being the theoretical value for the average number of contacts of jammed ellipses). Donev et al. [4] presented data in 3D on mono-size ellipsoids and spheroids obtained by a dynamic concurrent algorithm-based DEM. In both cases, the volume fraction at random dense packing of spheres of 0.64 increased to 0.71 for spheroids at an aspect ratio of about 0.6, and for ellipsoids to only a slightly higher volume fraction at an aspect ratio of about 1.5. Average number of contacts per particle increased in both cases from about 6 (spheres) to 10, approximately. All those simulations use periodic boundaries for exclusion of wall effects. Comparison with experiments in real containers (like those with M&Ms, Milk Chocolate Candies) is therefore not straightforward.

In our 2D simulations we also employed partly periodic boundaries. Loose as well as dense random states were explored (the latter through compaction on the top surface of the “container”). Fig. 6 shows the density differences in 2D for both regimes at different particle elongation (= reciprocal of aspect ratio) of the ellipses. The curve for compacted grains is quite similar to that in Donev et al. [4] on a slightly lower level. Average coordination number of circles is about 4 and somewhat higher for the ellipses in loose packing; in general lower than in Delaney et al. [3]. This is due to different algorithms as well as different particle samples being used. Delaney et al. [3] used a polydisperse particle size distribution to avoid the ordered structure, which could obviously have increased the coordination number.

A similar procedure but with the rigid boundaries is followed in the case of the 3D packing of mono-size standard polyhedra (with facet number 4 to 8) and an irregularly-shaped particle. 864 particles with sieve size 10 mm were used for each simulation. Fig. 7 visually illustrates some loose packed structures with several typical shapes. As facet number and sphericity are the sensitive parameter for these shapes, loose packing density is presented in Fig. 8 as function of these parameters. Sphericity is defined as surface area ratio of the equivalent sphere and the particle (both having equal volume). Loose random packing density is generally increased with increasing facet number and sphericity as similarly found in case

Fig. 6. Packing density of ellipses as a function of elongation (left) and frequency distribution for the coordination number of the circles as a function of the regime
Fig. 7. Mono-size random loose packing states of particles with some typical shapes.

Fig. 8. Random loose packing density as function of (left) facet number and (right) sphericity of particles.

of random dense packing [12]. Both cases of packing density have revealed particle shape as an extremely important factor in particle packing. It seems that polyhedra with larger sphericity can be packed to higher density. Similar tendencies are found for dense packing of the same standard mono-size polyhedra, shown in Fig. 9.

Coordination number is an important parameter for evaluating packing efficiency. As in 2D space, a numerical method is established for calculation of coordination number of a packed system of arbitrary-shaped particles. Certain additional mesh and evaluation points are applied to surface of particles for the assessment purpose. Its precision is related to the

Fig. 9. Random dense packing density of 3D tetrahedra and a spherical particle versus facet number (left) and as a function of sphericity (right).
Fig. 10. Frequency distributions of coordination number of dense randomly packed polyhedra of different types

fineness of the evaluation mesh. Selecting a coarse mesh speeds up the calculation, but leads to biased results. Therefore, a refined mesh should be applied for calculation of coordination number. The resulting distributions of coordination number with different shapes are presented in Fig. 10. So, coordination numbers in polyhedra packing are also not only related to packing density, but also related to shape.

POROSIMETRY IN MATURER STATE

Pore delineation stage
A random point system is superimposed on the virtual material. Only those points inside the pores are considered. A path planning algorithm inspired by the rapidly exploring random tree (RRT) approach developed in robotics 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 led to the name: Double Random Multiple-Tree Structuring (DRaMuTS). Details of the methodology are described in [13,14]. 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 $10^{-9}$ and $10^{-7}$ tree edges. Since the finest pores probably do not contribute significantly to the transport mechanism, a lower sensitivity level could be selected for practical purposes making the approach even more economic. Fig. 11 shows tree structures for PC and for gap-graded blended PC for all detected pores (left) and for the main trunks of the pore network connecting the outer surfaces of the specimen (right).

Pore measuring stage
The second stage starts by the aforementioned generation of the second random system of points of which the ones outside pore space are removed. The point fraction associated with the channels equals its volume fraction or continuous pore fraction. So, 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. 12 (left) convincingly demonstrates.

Fig. 12. Gradient structures of total porosity as influenced by gap-graded blending with RHA (left). Pores are coarser inside the ITZ zone in PC, however disproportionately refined by gap-graded blending (right). Note that MZ is middle zone, between ITZs; Pc=Portland cement; Po20 is Portland cement with 20% RHA; W40 = W/B or W/C=0.40)
Next, the random points are provided with a star, \textit{i.e.} from each point lines run in random or systematic directions to the nearest surface of the pore, forming the stars’ pikes. Length \( l \) of all pikes are measured per point, cubed and averaged. Twice the third root value is an estimate of the local pore diameter, \( d_r \). Hence, \( d_r = 2\sqrt[3]{\langle l^3 \rangle} \). The method is called star volume measuring and is developed basically for estimating pore size from 2D sections [15]. 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 (\textit{i.e.}, RHA) on the PoSD could be revealed this way (Fig. 12) [16]. With the eye on use of pore size characteristics 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 sections through a random point. A 2D star is defined in such a section by a random or systematic set of pikes in the section plane. Pike lengths \( l \) are determined per point, squared and averaged; cross section area is \( \pi l^2 \). 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 would be the major parameters in a transport model.

CONCLUSIONS

DEM by HADES is a reliable and economic way to simulate particle packing on both levels of the microstructure in concrete. For aggregate, at least the dense random packing state should be produced; for the binder, the loose packing state could be appropriate at low water to binder ratios representative for High Performance Concretes.

HADES renders possible simulating arbitrarily-shaped particles. For economic reasons, however, a limited number of standard shapes of ellipsoidal and polyhedron categories should be selected. The research dealing with shape effects on packing are qualitatively in agreement with physics studies in 2D and 3D. This approach therefore renders possible studying effects of aggregate grain shape (river gravel versus crushed rock) on packing density both in combination with variations in relevant technological parameters.

The non-spherical nature of cement grains can be captured in packing studies in the fresh state as well. The far more complicated hydration algorithms as compared to those for spherical grains have still to be developed. They will make it possible performing porosimetry studies on an even more realistic virtual representation of concrete.

The completed methodology of HADES-CemHydSim-DRaMuTS is available for elaborating in an economic way comprehensive studies on influences of technological parameters on geometrical and topological pore characteristics. HADES simulation will produce information superior in reliability as compared to such information coming from application of popular RSA systems [17].

The favorable strength experiences with gap-graded blending by RHA in real concrete [18] are supplemented in the present virtual experiments as to porosity. This will exert also positive effects on durability.
REFERENCES