Particulate structure and microstructure evolution of concrete investigated by DEM
Part 1: Aggregate and binder packing

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Experimental approaches in concrete technology are time-consuming, laborious and thus expensive. Developments in computer facilities render possible nowadays realistically simulating the particulate structure and microstructure of cementitious materials. For that purpose, discrete element methods (DEM) are required because of more correctly simulating particle dispersion. Dynamic DEMs like Delft-produced SPACE and HADES were developed and applied. The approaches are outlined and usage is illustrated by means of assessing particle size effects on packing density of aggregate. Shape simulation is accomplished for simple shape families like ellipsoids and polyhedra. This type of particles is used so far for the simulation of structures consisting of gravel, crushed rock or cement particles.

Keywords: Discrete element method, concrete, aggregate, shape effect, cement paste, packing

1 Introduction

Cementitious materials have complicated structural characteristics. This holds for the aggregate as well as for the hydraulic binder. Traditionally, large experimental programs have to be set up and executed for getting insight into the effects of technological variables that are of practical engineering interest on structural parameters and on material properties. This experimental route is obviously time-consuming and expensive. Since cementitious materials are of particulate nature, researchers can therefore also pursue considering structure parameters as resulting from particle packing experiments. An extensive amount of information on this topic is available in physics and mathematics literatures. So, starting from scratch is not necessary. Unfortunately, goals of the research underlying such publications are generally different from that in concrete technology. Moreover, most of the papers are dealing with 3D spherical (or 2D circular) particles (Cooper, 1988, Fu and Dekelbab, 2003). This is not necessarily a proper basis for studying aggregate or binder concepts in concrete technology. Hence, more realistic computer simulation systems are required for getting reliable information on particle packing phenomena relevant for aggregates and binders in concrete technology.

Popular packing systems in concrete technology are so called random sequential addition (RSA) ones. Herein, particles are *placed* on locations inside a container space controlled by a random generator, thereby proceeding from large to small (again, mostly spherical) grains. Occurring "overlap" of grains leads to rejection. Particle interference that is a crucial characteristic for production of cementitious materials is excluded. As we have described in an earlier issue of Heron (Stroeven and Stroeven, 2001), we therefore started *halfway the 1970s* with the development of a simulation system that can now be classified as a primitive version of a static discrete element method (DEM). The international publications recognized to have introduced DEM for the first time is from the *end of the 1970s* (Cundal and Strack, 1979). Halfway the 1980s we started developing a new version, which now could be classified as a dynamic DEM. It was among other things applied to the assessment of steel fibre efficiency in boundary layers of steel fibre reinforced concrete (SFRC) (Stroeven, 1986). Finally, halfway the 1990s the more sophisticated dynamic DEM with the acronym SPACE became available (Stroeven 1999). It was completed with a vector approach to cement hydration (Navi and Pignat, 1999; Jennings, 1986; van Breugel, 1991; Stroeven, 1999). In Stroeven and Stroeven (2001), the DEM system and its hydration components are described in detail and applications to the aggregate and the binder structure were provided. SPACE is restricted to spherical grains, as it is the case for most popular RSA systems in vogue in concrete technology. Dynamic features of SPACE are based on an impulse-based Newtonian mechanism for particle interaction, which leads to more realistic particle dispersion than can be achieved by the RSA systems, as demonstrated in the open literatures (Williams and Philipse, 2003; Stroeven *et al.,* 2009b). Fig. 1 shows the effect of particle dispersion on the de-percolation process. A higher degree of order in RSA systems (here represented by the Delft-produced HYMOSTRUC3D system (van Breugel, 1991)) and too much disorder in the digitized approach at NIST (Bentz *et al.,* 1994) lead to serious deviations in the de-percolation process during hydration. The DEM data are due to packing simulation by the SPACE system and 3D reconstruction after serial sectioning of the virtual cube of hydrated cement, thereby following Ye *et al.* (2003). Fig. 2 shows the much patchier characteristic of DEM dispersed (cement) particles.

During the first decade of the 21st century, two PhD studies explicitly focussed in more detail on the capillary pore network structure (Hu, 2004) and on the aggregate structure (He, 2010). In the latter case, a transfer to a new dynamic DEM system, *i.e.* HADES, was realized, which could incorporate also arbitrarily-shaped particles. This is highly relevant

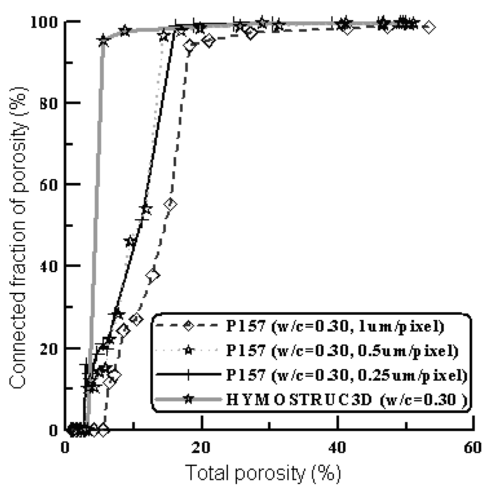
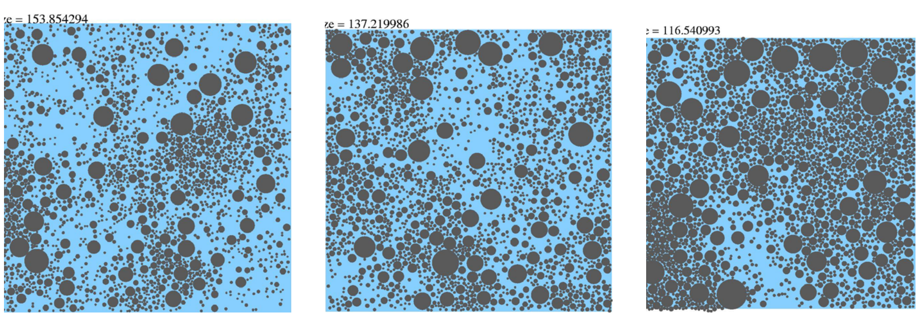


Figure 1: De-percolation process of simulated cement pastes analysed by RSA and DEM procedures



w/c=0.50 w/c=0.42 w/c=0.34

Figure 2: Sections of cubes with rigid boundaries of DEM-produced fresh cement paste models at increasing particle density. Material density is obviously fluctuating, despite particle attraction (or repulsion) not being included in the dynamic packing process by SPACE. As a result, "patches" can be distinguished (Diamond, 2006, Stroeven et al., 2009a).

for studying effects of the shape of aggregate (river aggregate versus crushed rock aggregate) on its packing density, with implications for concrete composition and strength properties. It may also be relevant for binder simulation. Neither particles of Portland cement (PC) nor ones of mineral admixtures are spherical, as experimentally demonstrated for PC by Garboczi and Bullard (2004). This will influence the packing structure in the fresh state of the paste as well as the hydration process, involving the pore de-percolation process as well as the resulting topological and geometric properties of the pore network structure (Bullard and Garboczi, 2006). The very complicated particle interferences for such hydrating systems are foreseen to be completed in a forthcoming PhD study. Hence, herein PC as well as mineral admixture particles are still considered spherical.

Prime goal of this publication (encompassing Part I and Part II) is to outline and discuss the features and capabilities of the present version of the dynamic DEM, HADES, which basically renders possible simulating the packing of artificially-shaped particles on different levels of concrete's structure, of the pore exploration system, DraMuTS, and of a novel and modern quantitative assessment methodology of pore topology and geometry, relevant for durability estimation. HADES will also be applied in a running PhD study for nano-level particle (globules) packing and porosimetry intended to more closely reflect latest observations on the hydrate structure (Muller *et al.*, 2013).

HADES is not unique, although it is one of the most versatile systems. More DEMs are used in concrete technology, mostly for specific purposes. For a survey of such systems, see (Stroeven *et al.*, 2009b).

2 Dynamic discrete element method for particle packing (HADES)

The particles that should be dispersed in a container are initially dispersed with a random generator in a much larger container with similar boundary conditions. Properties of this ensemble of particles are derived from the sieve curve when aggregate is concerned, or from the so called Rosin-Rammler distribution function (Hu, 2004)

$$f(d) = 1 - \exp(-bd^{n}) \tag{1}$$

when the cement in the binder is at issue. In Eq. (1), b and n are constants. The particles have a shape representative for the aggregate or binder being simulated. Of course, the economy of the experiments does not allow giving each particle an individual shape. Instead, families of elementary shapes have been implemented in HADES, such as ellipsoids and polyhedrons (He, 2010). Mixtures can be produced of types of polyhedrons or of ellipsoids to yield optimum simulation conditions with respect to the real particle mixture. In He *et al.* (2012) a mixture of nine regular polyhedrons is designed with the same global shape parameter (involving surface area and volume of particles) as experimentally assessed for four different aggregates by Erdogan *et al.* (2005). For the cement particles also the global surface to volume ratio should be a leading parameter, because it will significantly influence the packed particle structure in the fresh state as well as the hydration process. As a consequence, it will govern the characteristics of the resulting pore network structure.

Next, the dispersed particle ensemble is subjected to the dynamic stage. Particles are set to move and rotate linearly in a random way as was also realized in SPACE. Detection of near-collisions between grains or between grains and the container boundaries are solved differently for non-spherical grains, however. In HADES, the approaching surfaces of grains (or container boundary) are locally tessellated. Moreover, thin fictitious guard zones of constant thickness T are stretching over particle and container surfaces. During the approach, all elements activated by overlap of mutual guard zones will contribute to increasing repulsive forces and moments between the grains (or grain and container boundary) that prevent real collision. Instead, the impulses on the particle force it to move and rotate along a new trajectory. Fig. 3 represents the situation of two particles that move and rotate in container space.

When the desired aggregate volume fraction or water to cement ratio of the paste is obtained, the simulation procedure is stopped. The container can be provided with rigid surfaces, with periodic ones, or with mixed conditions. In the first case, the simulated material forms boundary zones in the container. These are denoted interfacial transition zones (ITZs) in case that the simulated material is the hydrated binder material. The rigid surfaces are representing those of the aggregate. The effect of nearby boundaries is eliminated when periodic boundary conditions are used. The result is a representation of so called bulk material. Cement paste pocketed by only two nearby aggregate grains is simulated in a container with two rigid and four periodic boundaries. This is realized in a container with six periodic boundaries and an internal sheet. When the bottom part of the simulated material is placed on top of the top part, the resulting material has two rigid and four periodic boundaries, as illustrated in Fig. 4.

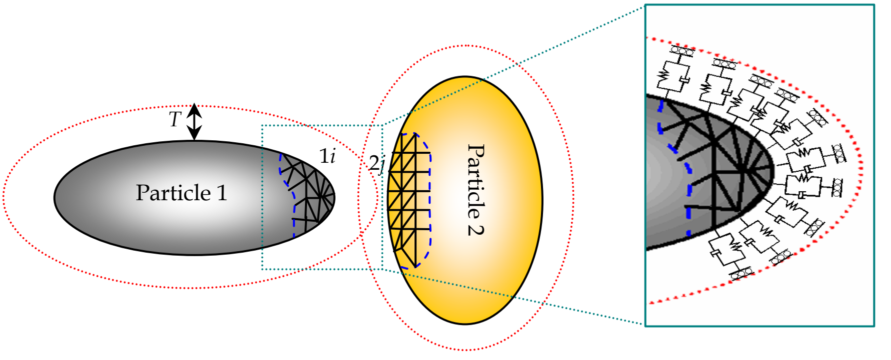


Figure 3: Mechanism in the dynamic stage to prevent particle overlap. Upon close encounter overlapping guard zones with thickness T locally activate tessellated surface elements that generate repulsive forces the intensity of which will depend on local spacing.

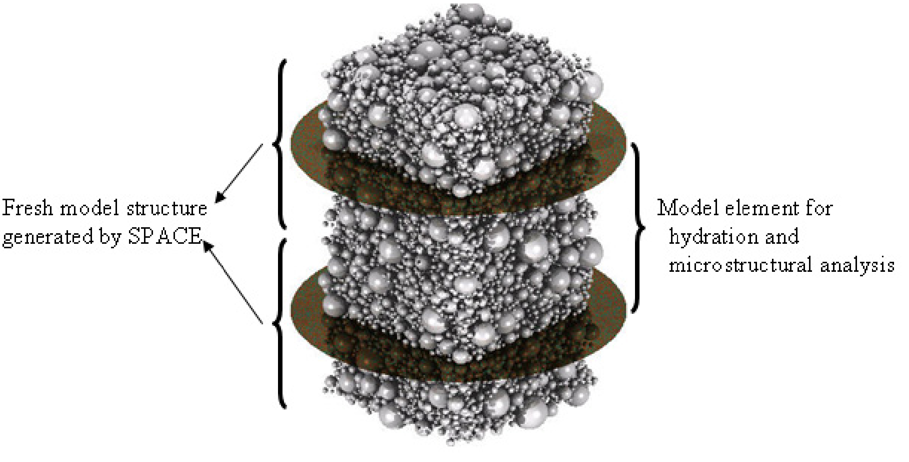


Figure 4: Fresh model cement paste structure with semi-rigid boundary condition.

3 Simulation of aggregate packing

In Stroeven and Stroeven (2001), it was already demonstrated that SPACE-simulated (spherical) aggregate mixtures complying with six different sieve curves and with particles of fluvial origin ranging from 1 to 32 mm could be packed to densities satisfactory close to experimental values obtained by standardized aggregate packing in 8 liter cylindrical containers. Thereupon, shape effects have been extensively studied for loose random and dense random packed aggregate by applying the HADES system (He *et al.*, 2009). Fig. 5 shows the basic simulation concept for river gravel (ellipsoids) and crushed rock aggregate (polyhedrons), respectively. A specific group of shapes with 4-8 facetted surfaces have been developed to represent practical crushed rock mixtures, following Guo's field investigations (Guo, 1988). This greatly simplifies the simulation of crushed rock since only two parameters, *i.e.*, sieve size and the maximum size of the surface element, are required in DEM. Fig. 6 displays the particles with different number of facets and surface tessellations.

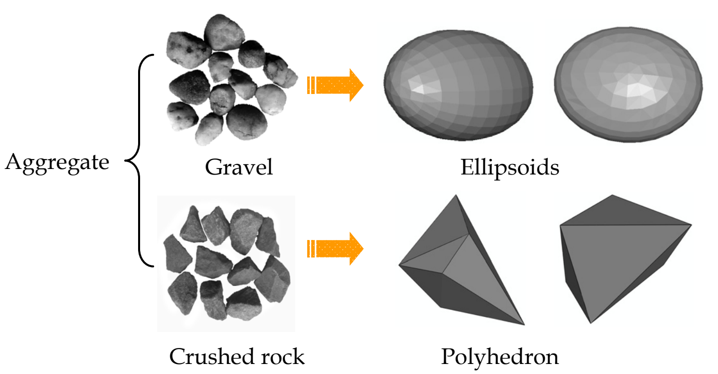


Figure 5: Representation of river gravel and crushed rock aggregates by ellipsoids and polyhedrons, respectively

As stated above, polyhedron shapes are all regular. As an alternative, arbitrary-shaped particles can be simulated by quantitative image analysis: particle shape is characterized by the vertical and horizontal profiles of the aggregate. By combining the information of two profiles by a 3D mesh program, the rough shape of real aggregate can be reconstructed. Fig. 7 shows an application on an arbitrary shaped aggregate. Of course, more sophisticated particle simulation approaches are available (Garboczi, 2002); however, the final result is generally only used as reference for the more economic polyhedron concept.

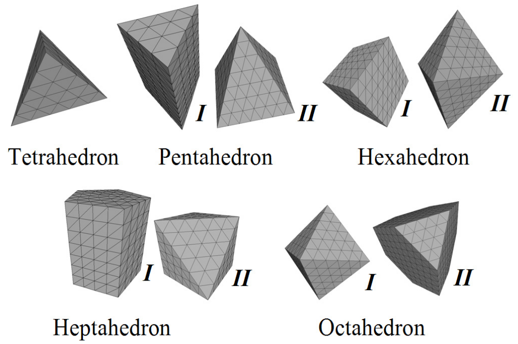


Figure 6: Nine regular polyhedra with facet number 4~8.

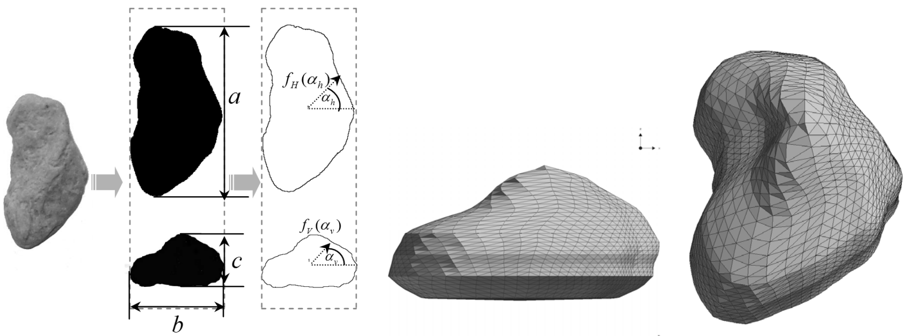
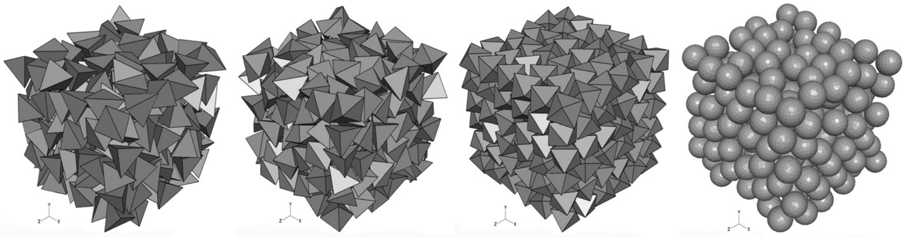


Figure 7: (left) Characterization of shape information of a real aggregate by image analysis method; (right) two views of the reconstructed corresponding grain

Next, Figs. 8-10 present results obtained on crushed rock aggregate simulations by HADES, which involved mono-sized particle packing, rendering possible comparison with packing results available in physics and mathematics (Donev *et al.*, 2004; Delaney *et al.*, 2005; Stroeven and He, 2013). In general, these simulations yield somewhat higher density and average coordination number because of employing different packing algorithms, *i.e.* particularly the guard zone is missing. However, in concrete technology this reflects the effect of the binder paste around the aggregate. Presented data reveal packing density significantly influenced by particle shapes. Packing density seems increasing with facet number and sphericity, whereby sphericity is defined as the surface area ratio of the equivalent sphere with that of the particle (having equal volume). The same tendency was also found for loose random packing states. Similar findings were reported by German

(1989). Note that particles can be additionally provided with a certain degree of roughness, too (He, 2010). This will exert influence on the packing structure, of course.



(a) (b) (c) (d)

Figure 8: Mono-sized simulated random packing with particles having typical shapes: (a) tetrahedron (b) hexahedron (c) octahedron and (d) sphere, respectively.

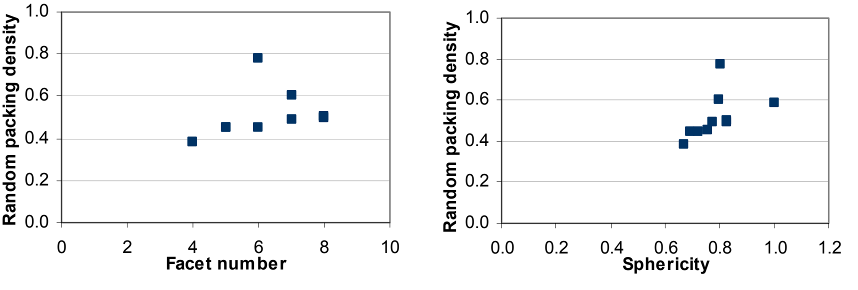


Figure 9: Dependency of maximum packing density on (left) facet number and (right) sphericity of 3D particles such as the ones shown in Fig. 8 (full list presented in Fig. 10).

4 Simulation of fresh binder packing

Modern high performance concretes are produced at low water to cement ratio. The volume content of the cement in the paste may be as high as 60%. The advanced DEM system HADES has been developed with capabilities of packing arbitrary shaped particles into the relevant dense random packing state (He *et al.*, 2009). A comparison study has been performed for finding out the preferable simplified shapes for simulation of cement particles (He *et al.*, 2012), in which the experimental results by X-ray tomography method are used as references (Garboczi and Bullard, 2004). The preferred shapes defined in that study can also be incorporated in HADES. After assessment of grain shape, mass properties are assigned to each particle, *e.g.*, mass, center of mass, moment of inertia.

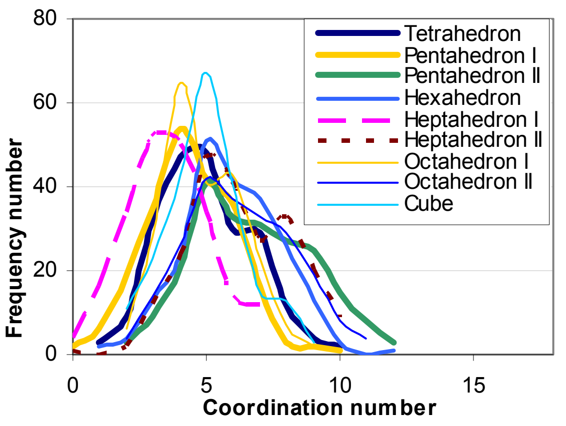


Figure 10: Frequency distributions of coordination number of dense packed polyhedrons of different types (This figure is in colour at www.heronjournal.nl)

Fig. 11 (left) presents *S-V* information of a group of simulated cement grains using flat ellipsoids, which fits experimental results (Garboczi and Bullard, 2004) well. A visualized structure of correspondingly loose packed ellipsoids obtained by HADES is plotted in Fig. 11 (right). Alternatively, Figure 12 (left) shows the *S-V* distributions of a randomly generated octahedrons example with an experimental reference curve. The figure reveals the cases of 1000 particles with longest axis in 10~50 μm size range packed by HADES. The *S-V*-relationship complies well with experiments. Fig. 12 (right) shows an example of a loose packed structure of arbitrary octahedrons, supposedly representing a fresh cement structure.

Morphological comparisons can additionally be made between sections of simulated particle structures and of real ones. As an example, Fig. 13 shows a random section of the structure in Fig.12 (right) and a 2D section of a real cement structure obtained by X-ray micro-tomography. Cement particles in Fig. 13 (right), displayed in light grey, obviously reveal the angular shape that is similarly revealed by the section of the polyhedral cement particles in Fig. 13 (left). This adds to the aforementioned quantitative matching of *S-V* features of real and simulated cement structures, as shown by Fig. 12 (left). Therefore, it can be concluded that the simulation by polyhedrons provides the more realistic solution for the given cement. In general, this approach by HADES will provide a significantly improved option for modelling of cement particles as compared to the conventional sphere system. This will also have impact on the hydration process, of course.

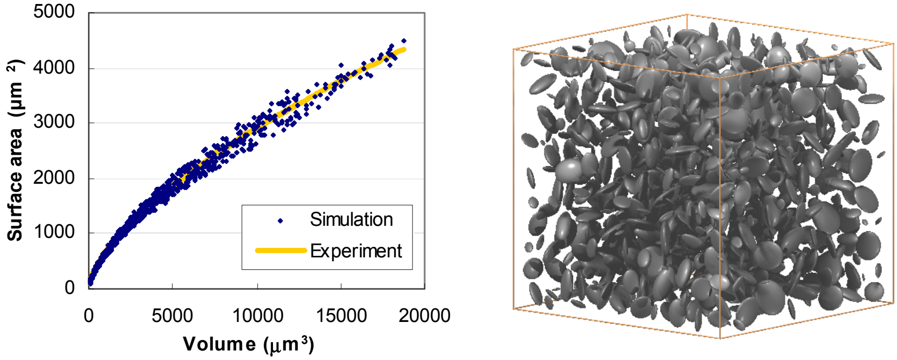


Figure 11: (left) S-V-relationships of a group of cement grains by flat ellipsoids with experimental regression results; (right) visualized structure of compacted grains by HADES (1000 particles with longest axis in 10~50 µm range)

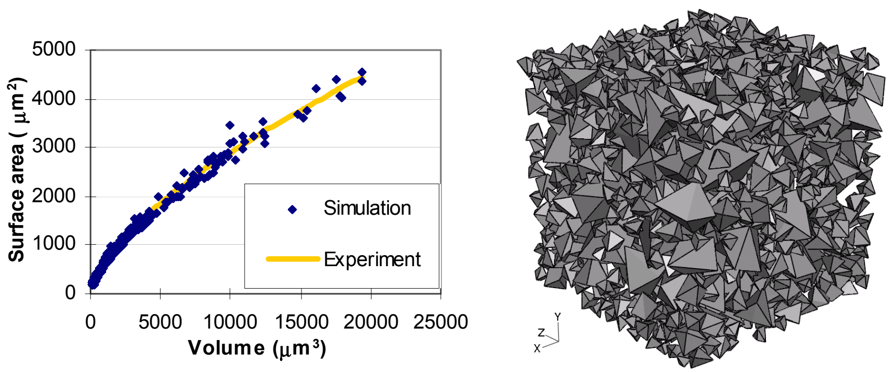
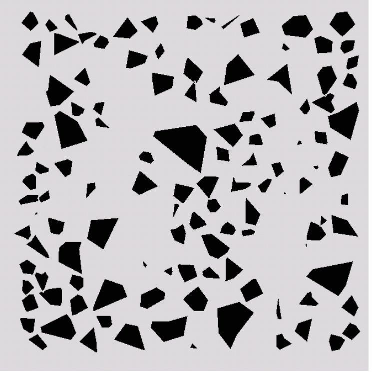


Figure 12: (left) Computer simulation of 1000 particles in the 10~50 µm size range and experimental regression results (Garboczi and Bullard, 2004) and (right) visualized structure of compacted grains by HADES

5 Conclusions

Fresh particle packing is crucially important for the granular structure formation and structural properties of concrete. DEM renders an economic and efficient approach to the properties assessment of particle packing. The paper presents the basic mechanisms as well as the capabilities of the developed DEM, i.e. HADES, on particle packing of concrete. With the discretized tessellated surfaces, simulation of basically any arbitrary shaped particle is possible by this system. However, simulating packed structures of such particles will not offer economic solutions. Therefore, simplified simulation strategies for aggregate and



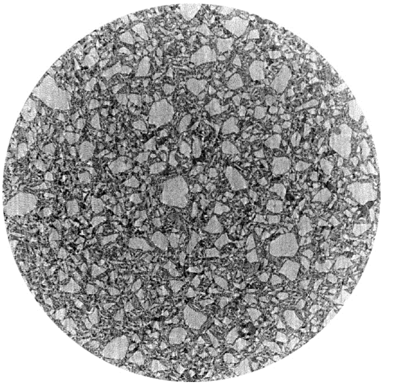


Figure 13: (left) section of the simulated structure (1000 octahedron grains in 10~50 μm range); (right) 2D section of a real fresh cement structure (Cement-133) obtained by X-ray micro-tomography; w/c=0.35. Source: http://visiblecement.nist.gov/cement.html

cement particle are set up based on comparisons with experimental results. Surface to volume information of aggregate and cement particles are emphasized by this method as it is important for the fresh meso-structural properties as well as microstructure evolution. Following this methodology, shape simulation strategies could be successfully implemented in this new DEM system. Applications on particle packing have revealed the typical shape effects on packing properties also known from physics and mathematics literature. As a consequence, more realistic materials structures on meso-level as well as on micro-level can be reconstructed with the current packing approach. Porosity properties can then be characterized for the hydrating packed binders. This is discussed in the second part of the paper.

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