Granular Matter (2008) 10:329–334
DOI 10.1007/s10035-008-0092-4

Characterization of voids in spherical particle systems by Delaunay empty spheres

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Received: 23 December 2006 / Published online: 29 February 2008

Abstract The pore space of mono-sized spherical particle systems of increasing density is characterized by Delaunay empty spheres. Periodic packings of densities ranging from 0.57 up to 0.70 are generated numerically by symmetric vibration. The Voronoi diagrams of these packings are then computed with an algorithm based on the research of Delaunay empty spheres. The voids distribution and the tortuosity of packings as a function of density are studied. As the density increases, the voids distribution becomes more narrow. For partly ordered packings of high density, the voids distribution presents two peaks corresponding to the size of Delaunay empty spheres of perturbed fcc or hcp packings. The tortuosity of disordered packings decreases slowly with density. However, when the system becomes partly ordered, a large increase in tortuosity is observed.

Keywords Packing · Delaunay empty sphere · Voronoi diagram · Voids distribution · Tortuosity

1 Introduction

The pore space characterization of granular materials is of great interest in a lot of industrial applications. In many cases, the pore size distribution, the connectivity and the tortuosity of pores are key parameters for the optimization of industrial processes and of material performances. However, the experimental characterization of the pore space of granular materials is relatively heavy and in some cases impossible. Simulation by numerical modelling can thus be very useful for the characterization of the pore space between particles [1–4] as well as for the characterization of the geometry of the granular system itself [5–7].

The objective of this paper is to study the pore space of mono-sized spherical particle modelled systems of increasing density, leading to random or partly ordered packings. The methods used to produce the packings and to compute Voronoi diagrams and Delaunay empty spheres are first briefly described. The distribution of pores as a function of the density of the systems is then studied. Finally, the Voronoi diagram and Delaunay empty spheres are used to give a simplified geometrical representation of the pore space, and to compute the tortuosity of the packings.

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2 Numerical procedures

2.1 Generation of periodic mono-sized spherical particle packings

Disordered packings of densities varying from 0.57, corresponding to a random loose packing [8,9] up to 0.644, corresponding to a random close packing [9–11] and partly ordered packings of densities varying from 0.635 up to 0.696 have been generated by symmetric vibration [12]. At first, an initial very loose granular system composed of 10,000 spheres of radius R is generated by Random Sequential Addition [13]. Particles are introduced one by one into a parallelepipedic container of dimensions $40R \times 40R \times 73R$ with periodic boundary conditions for the six faces. For each particle, a

Table 1 Denomination of the packings used in the study and corresponding densities

	Disordered packings				Partly ordered packings			
Packing	DP0	DP1	DP2	DP3	POP0	POP1	POP2	POP3
Density	0.570	0.605	0.630	0.644	0.635	0.662	0.689	0.696

random location is generated until it can be placed into the container without overlapping already placed particles. With this procedure, only low densities can be reached and the system cannot be considered as a packing because particles are not systematically in contact. This system is then densified by the mean of a sedimentation procedure respecting the periodic boundary conditions, in order to produce a stable random loose packing of density 0.57. This packing, called DP0, is then submitted to a series of vibration cycles of amplitude α. For each cycle, the particles are chosen in a random order and attempt to perform an upwards vertical displacement of length α. If this displacement leads to an overlap with another particle it is rejected, and up to four random lateral upwards displacements of length lower or equal to α are generated until the particle can be placed. If all these displacement attempts lead to overlaps, the particle remains at its initial place. Then, similar downwards displacements are imposed to the particles in a new random order. The detailed description of the rules used for the displacements of particles is presented in [14]. It has been shown that, under these conditions, the system can be transformed into a "suspension" in which a proportion of particles have the possibility to vibrate symmetrically [12,14,15]. The system can then be densified with the former sedimentation procedure. By varying the amplitude of vibration α and the frequency of sedimentation procedures, a wide range of disordered or partly ordered packings can be generated. The complete description of the model used and the simulations performed for the generation of these packings are presented in details in a previous work [12]. Four disordered packings called DP0, DP1, DP2 and DP3 and four partly ordered packings called POP0, POP1, POP2 and POP3 have been produced. Table 1 presents the different packings and their corresponding densities.

2.2 Computation of Voronoi diagrams and Delaunay empty spheres

2.2.1 Definitions and properties

$\Sigma = \{P_1, P_2, \dots P_N\}$ being a set of spherical particles, the Voronoi region VR_i of particle P_i is defined as the locus of all the points closer to the surface of P_i than to the surface of any other particle $P_j (j \neq i)$. The ensemble of all the Voronoi regions is the Voronoi diagram of Σ.

Each Voronoi region is bounded by a series of faces f_{ij}, which are the locus of all the points equidistant to the surface of P_i and P_j and closer to the surface of P_i and P_j than to the surface of any other particle $P_k (k \neq i, j)$. Two particles P_i and P_j sharing a face f_{ij} are neighbours (Fig. 1a).

Each face f_{ij} is limited by several edges $e_{i,j,k}$, locus of all the points equidistant to the surface of at least three particles P_i, P_j and P_k, and closer to the surface of these particles than to the surface of any other particle $P_l (1 \neq i, j, k)$ (Fig. 1b).

The extremities of each edge $e_{i,j,k}$, correspond to two vertices $v_{i,j,k,l}$ and $v_{i,j,k,m}$ which are equidistant to the surface of at least four particles P_i, P_j, P_k and P_l (or P_m) and closer to the surface of these particles than to the surface of any other particle (Fig. 1c).

In general, an edge is equidistant to exactly three particles and a vertex to exactly four particles. However in some cases, particularly in ordered systems, an edge can be equidistant to more than three particles and a vertex can be equidistant to more than four particles. These edges and vertices are called degenerated.

According to the previous definitions, each vertex $v_{i,j,k,l}$ of the Voronoi diagram defines the centre of a sphere tangent to at least four particles P_i, P_j, P_k and P_l without overlapping any other particle of the system (Fig. 1d). This sphere, called Delaunay empty sphere [16], is inscribed into the pore space between particles. The vertices of the Voronoi diagram can be determined by the position of Delaunay empty spheres and vice versa.

2.2.2 Method used

The algorithm used in this study for the computation of the Voronoi diagram of a granular system is based on the research of the positions and radii of Delaunay empty spheres. The complete determination of the Voronoi region VR_i of a particle P_i is performed as follows:

1 An empty sphere of very small radius, tangent to the particle P_i is first seeded in the porosity.
2 The radius of the empty sphere is increased with an increment ε and its position is changed until it becomes tangent to three particles, P_i, P_j and P_k. In this position, the empty sphere is located on the Voronoi edge $e_{i,j,k}$.
3 The empty sphere is moved along $e_{i,j,k}$, (its radius is changed so that is remains always tangent to P_i, P_j and P_k) until it becomes tangent to a fourth particle P_m. In this later position, the empty sphere is located on the Voronoi vertex $v_{i,j,k,m}$.
4 Step 3 is repeated for each Voronoi edge of VR_i until all the Voronoi edges have been traced. The whole Voronoi region VR_i is then completely determined.

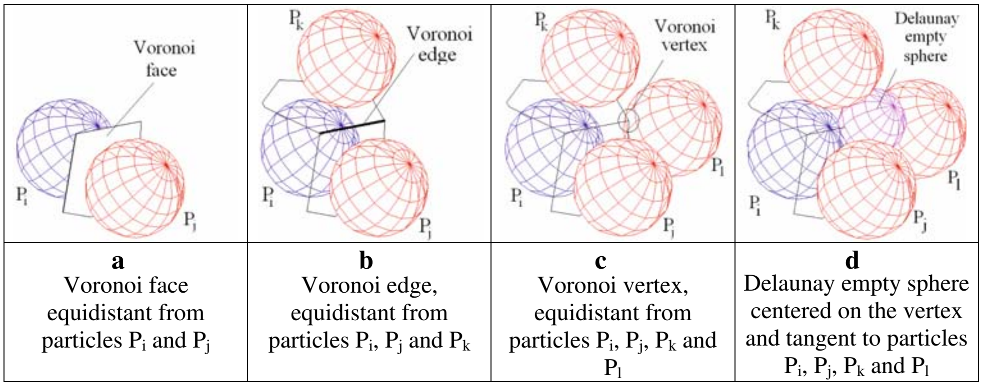

Fig. 1 **a** Voronoi face equidistant from particles P_i and P_j. **b** Voronoi edge, equidistant from particles P_i, P_j and P_k. **c** Voronoi vertex, equidistant from particles P_i, P_j, P_k and P_l. **d** Delaunay empty sphere centred on the vertex and tangent to particles P_i, P_j, P_k and P_l

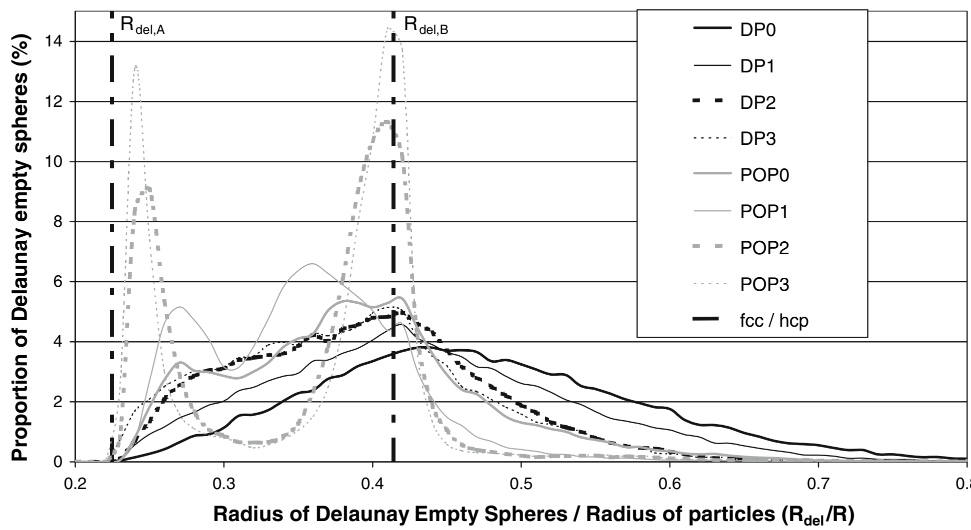

Fig. 2 Delaunay empty spheres radii distributions of the different packings

The tracing of Voronoi edges for the computation of Voronoi diagrams has already been used by Luchnikov et al. [17] and by Kim et al. [18]. Its main advantage is that it can be generalized to poly-disperse particle systems composed of spherical or nonspherical particles. Moreover, by recording the positions and radii of empty spheres moving along Voronoi edges, it is possible to describe the pore space of the granular system as a set of spheres inscribed into the porosity.

3 Delaunay empty spheres radii distributions

Figure 2 presents the distributions of Delaunay empty spheres radii (R_{del}) of the studied packings. Two kinds of Delaunay empty spheres radii distributions can be distinguished, depending on the geometry of the packings. On one hand, the distributions of disordered packings (DP0 to DP3) are very spread, the mean value of Delaunay empty spheres radii decreasing with the increase in density of the packings ($R_{\mathrm{del,mean}} = 0.469R$, $0.429R$, $0.389R$ and $0.380R$ for DP0, DP1, DP2 and DP3 respectively). On the other hand, the distributions of partly ordered packings (POP0 to POP3) are more narrow and present two peaks, the height of which increases with density.

The previous results can be explained considering the Delaunay empty spheres characteristics of perfectly ordered face centred cubic (fcc) or hexagonal (hcp) mono-sized sphere systems. These two systems have a density of 0.7405, and are the densest packings that can be realized with mono-sized spheres. The characteristics of their Delaunay empty spheres are identical, two kinds of Delaunay empty spheres coexist (Fig. 3). The smaller Delaunay empty spheres (type-A, $R_{\mathrm{del,A}} \approx 0.2247R$) are in contact with 4 particles and each Voronoi region contains 8 vertices of this type. The larger empty spheres (type-B, $R_{\mathrm{del,B}} \approx 0.4142R$) are in contact with 6 particles and each Voronoi region contains 6 vertices of this degenerated type. The Voronoi diagram of a perfect fcc or hcp packing has therefore to contain a proportion of type-A vertices equals to 2/3. The denominations type-A and type-B

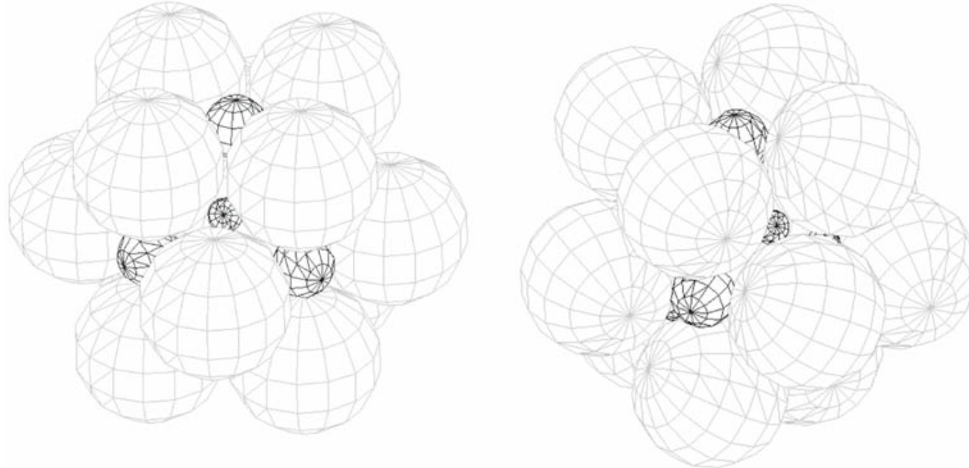

Fig. 3 Views of the neighbours (*light spheres*) of a particle in fcc (*left*) and hcp (*right*) packings and of the Delaunay empty spheres (*dark spheres*) corresponding to the Voronoi Region of the particle

vertices refer to the denomination of topologically stable and nonstable vertices given by Troadec et al. [19]. These authors have shown that for fcc or hcp packings, a small perturbation removes the degeneracy of type-B vertices by creating 4 new vertices, each new vertex being tangent to only four particles.

The partly ordered packings can be regarded as perturbed ordered packings presenting an increasing perturbation from POP3 to POP0. So the Delaunay empty spheres radii distribution of packing POP3 (partly ordered packing of highest density) presents two peaks very close to perfectly ordered fcc or hcp packings radii $R_{\mathrm{del,A}}$ and $R_{\mathrm{del,B}}$. However, the second peak is almost centred on $R_{\mathrm{del,B}}$ whereas the first peak has clearly shifted towards larger radii. Type-A Delaunay empty spheres correspond in fact to the smallest possible Delaunay empty spheres in a mono-sized sphere system. In a partly ordered packing, the positions of particles do not correspond exactly to the theoretical positions of the crystalline lattice and particles are not as close as in the perfect crystal. The smaller voids are therefore larger than those in the crystal, which explains the shift of the first peak towards larger radii. The second peak corresponds to Delaunay empty spheres of type-B. In this case, the perturbation leads to the removal of degeneracy of these vertices [19]. In the perfect crystal, type-B Delaunay empty spheres are tangent to exactly 6 particles. If the perturbation brings a particle away from the centre of the corresponding Delaunay empty sphere, new empty spheres will be created, but their radii will be very close to the initial value. However, if the perturbation brings a particle closer to the centre of the Delaunay empty sphere, the radii of empty spheres will slightly decrease. This explains the shift of the second peak towards smaller radii in the case of stronger perturbations (POP1 and POP0). For POP1 and POP0, the ordered parts of the system are surrounded by large disordered regions which explains the presence of a third peak resulting from the superimposition of the Delaunay empty spheres radii of the ordered parts and those of the disordered parts of the packings.

In disordered packings (DP0 to DP3), no characteristic peak can be observed in the Delaunay empty spheres radii distributions. The increase in density leads to a diminution of Delaunay empty spheres radii and of the spreading of the distribution. Due to the existence of a lower bound for Delaunay empty spheres radii ($R_{\mathrm{del,A}}$) the distribution shows a "swelling" for radii smaller than $0.3R$.

4 Description of pores with Delaunay empty spheres

Voronoi diagrams can also be used to give a simplified geometrical description of connected pores. As a matter of fact, Delaunay empty spheres represent the larger spherical pores of the packing, which can be connected by Voronoi edges. A pore linking two connected Delaunay empty spheres can be described by an empty sphere of varying radius moving along the corresponding Voronoi edge. Starting from the first Delaunay empty sphere, the radius of the moving empty sphere decreases until reaching a minimum value corresponding to the radius of the "bottleneck" of the pore. Then, its radius increases until reaching the radius of the second Delaunay empty sphere. The shape of this pore can therefore be represented approximately by two truncated cones linking the two Delaunay empty spheres to the "bottleneck". We have used this simplified geometrical description in order to represent some pores of the studied packings.

Moreover, in order to characterize the tortuosity of the studied packings, the more direct pathways crossing the packings from bottom to top have been followed. For each

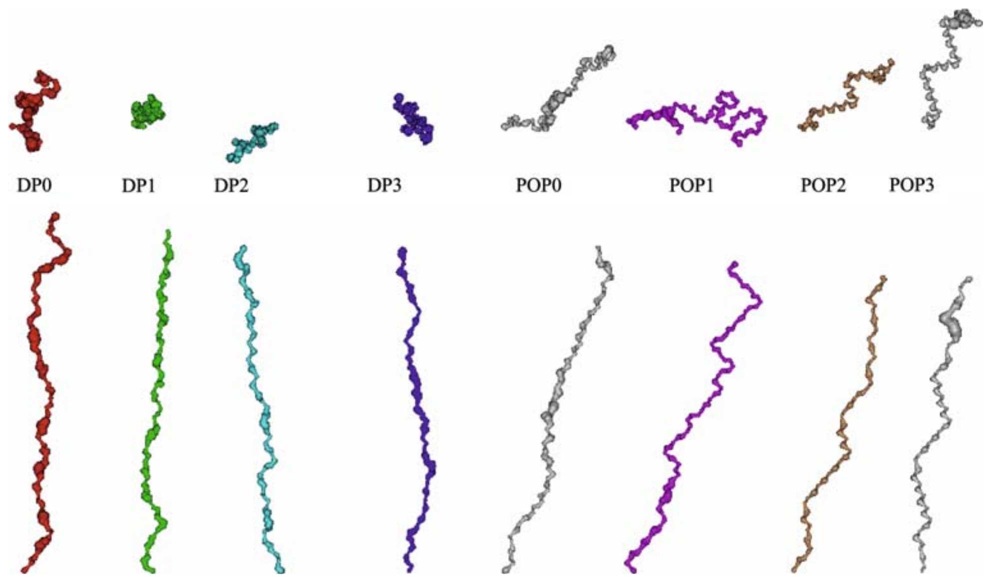

Fig. 4 Views (*top and face*) of typical connected pathways in the packings

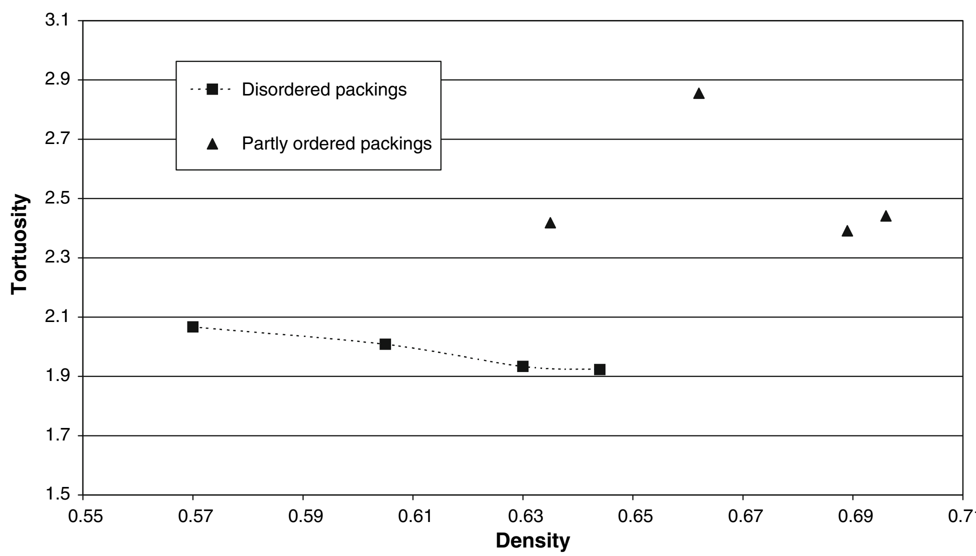

Fig. 5 Variation of the mean tortuosity as a function of the packing density

packing, all the Voronoi vertices have been scanned. Each vertex of which the corresponding Delaunay empty sphere straddles the bottom of the packing is recorded as the beginning of a connected pathway. Then, all the Voronoi edges connected to this Delaunay empty sphere are scanned and the more vertical edge, leading to a Delaunay empty sphere located above the former is followed. By repeating this procedure until reaching the top of the packing, a connected pathway crossing the whole packing can be drawn. The tortuosity τ of each connected pathway has then been computed as following: $\tau = (L/H)^2$, where L is the total length of the Voronoi edges that have been followed, and H the total height of the packing. The mean tortuosity of the packing has finally been computed by averaging the tortuosities of all the connected pathways.

Figure 4 presents views (top and face) of a connected pathway drawn for each studied packing. Figure 5 presents the variation of the mean tortuosity of the packings as a function of density for disordered and partly ordered packings. These two figures clearly show that the tortuosity of partly ordered packings is much larger than that of disordered packings. For the latter, the tortuosity decreases slowly with density. On the contrary, for partly ordered packings the variation of the mean tortuosity is more complex. The ordered parts of the systems lead indeed to very inclined pores and therefore to a higher tortuosity. However, it has to be pointed out that tortuosity is computed along the height of the packings (the more vertical edges leading from one Delaunay empty sphere to another are selected). In partly ordered packings, tortuosities computed along the principal directions of the crystalline lattice would be smaller.

5 Conclusion

The computation of Voronoi diagrams and Delaunay empty spheres is a very efficient tool for the study of the pore space of modelled packings as it allows to derive the pore size distribution as a well as the connectivity and tortuosity of pores.

The study of the pore space of mono-sized spherical particle systems of increasing density shows that the voids distribution of disordered packings is spread, the mean radius and the spreading of the distribution decreasing with the increase in density. On the contrary, the voids distribution of partly ordered packings presents two peaks, corresponding to the radii of Delaunay empty spheres of perturbed fcc or hcp packings.

The simplified geometrical description of pores shows that connected pathways crossing disordered packings from bottom to top are rather vertical whereas they are very inclined in partly ordered packings leading to higher tortuosities.

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