Three-Dimensional Apollonian Packing as a Model for Dense Granular Systems

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An iterative procedure according to which new spheres are inserted between existing ones is realized for computer simulation of dense granular systems. The method is based on the rigorous geometrical Voronoi-Delaunay approach generalized for pore volume analysis inside an ensemble of polydisperse hard spheres [N. N. Medvedev, Dokl. Akad. Nauk **337**, 767 (1994) [Dokl. Phys. Chem. **337**, 157 (1994)]]. Models with packing fraction $\eta \sim 0.9$ are constructed. The particle size distribution and fractal nature of the packings are investigated. The fractal dimension $D = 2.45$ is obtained.

The structure of the dense packings of hard spherical particles is an old but still interesting problem. The general laws of space filling in noncrystalline systems are of basic interest for researchers; for industry it is important to obtain packings with high bulk density. Therefore the packing of hard spheres is an object of permanent experimental and theoretical studies (see, e.g., [1–11]).

The bulk density of packing is characterized by packing fraction η, or porosity $\varepsilon = 1 - \eta$ being a fraction of unoccupied volume. Different factors have effects on packing density. However, the particle size distribution is known to play a basic role. The maximum value of η for random packings of equal hard spheres amounts up to about 0.64 [2–4]. More dense packings can be obtained using spheres of different sizes by filling the gaps between large particles with the smaller ones. In this case, theoretically, any value η close to unity can be reached. However, many small particles should be involved, and the largest and smallest radii will differ by many orders of magnitude. In practice, the range of particle radii is always limited. The size of coal particles used to produce grinds for coke making differ by a few orders [5]. The difference is less for ceramic and metallic powders. For atomic systems the atom radii differ no more than by a factor of 2–3. There are analytical approaches that study relations between a particle composition and bulk density for binary and multicomponent mixtures [6,7]. However, the general problems of hard sphere packings are far from being solved.

Computer simulation of packings is especially interesting. In this case, the radii and coordinates of particle centers are available. There are many methods to construct computer models of hard sphere packings. Some of them were considered in the review [8] reporting the packings of equal spheres and binary mixtures. Other particle size distributions including the Gaussian and log-normal laws were used in [9] and [10]. However, it is not so easy to obtain a high packing density in computer simulation. The authors of these papers failed to get η value exceeding 0.65 for their polydisperse systems using the conventional Monte Carlo [9] or the "Bennet-type" methods [8,10].

In this Letter we are proposing a geometrical method for constructing packings of any density by performing a procedure according to which the new spheres best fit between the existing ones. For a two-dimensional case such a packing is called the Apollonian packing [12] after the ancient Greek mathematician who had considered the problem of the circle inscribed among three given circles in a plane. For our purposes, however, we must be able to solve a three-dimensional Apollonian problem, i.e., to inscribe a sphere among four given spheres in a space. Recently, such a problem was solved at the generalization of the well-known Voronoi-Delaunay approach to the analysis of unoccupied volume distribution in the ensemble of differently sized spheres [13].

The Voronoi-Delaunay approach has long been employed to study the structure of the computer models of simple liquids and amorphous phase, see, e.g., [14]. In the last years, researchers have used this method to describe the unoccupied volume inside packings of equal spheres. The pore size distribution was studied using Voronoi polyhedra [15] and Delaunay simplices [16]. The Voronoi network was also used to study the permeability of the dense packings of equal spheres [17].

We are omitting here the details of the Voronoi-Delaunay generalization to nonequal spheres. Note, however, that the known algorithms for Voronoi polyhedra and Delaunay simplices [18,19] does not work for this case. The usual Voronoi polyhedra fail in studies of the volume between particles [20]. Indeed, the Voronoi polyhedron is determined for a particle center and is the volume of a region all points of which are closer to the given *center* than to the *centers* of all other particles in the packing. Studying the unoccupied volume, which is limited by particle surfaces, one must deal with another construction. It can be called *the Voronoi S region* [13] and defined as the region of volume all points of which are closer to the *surface* of the given sphere than to the *surfaces* of other spheres of the packing. Both of these constructions are identical for systems of equal spheres, but for different radii they are diverse. However, if there is not a large difference in sphere radii, or the packing is rather

    0031-9007/95/75(23)/4314(4)$06.00

dense and homogeneous, the Voronoi S regions resemble the usual Voronoi polyhedra. They differ only in the fact that Voronoi S regions have slightly curved edges and either convex or concave faces (see Fig. 1 in [13]).

The Voronoi-Delaunay method is convenient for our purposes, because the vertices of the Voronoi S regions (as well as those of the Voronoi polyhedra in monodisperse systems) determine the centers of maximum sized spheres inscribed between particles. Having at hand these interstitial spheres for a given packing, one can turn some of them into new particles. Avoiding overlapping between the new particles, we get a new configuration of hard spheres. One can exploit this technique in an iterative loop to create a more and more dense packing (see Fig. 1).

Our models were constructed in a cube with periodical boundary conditions. The largest number of particles we could create in a packing was about 40 000. This allows us to reach the value $\eta \sim 0.9$ starting with the initial configurations consisting of about 100 particles. This limit is not a restriction of the method, but a result of our computer power. The main CPU time and RAM are needed when calculating the Voronoi S-region tessellation to get the whole set of interstitial spheres.

The model systems discussed below were constructed to fill in the biggest pores first. The interstitial spheres with radii exceeding a given cutoff length r_c were taken into account. The spheres with the largest radii were turned

into new particles first. We had to perform a few iterations (i.e., put new particles into the packing and recalculate the interstitial spheres) to fill in all the gaps in the packing where the particles of radius equal to r_c or larger can be inscribed. Thereafter the current value of the cutoff length was decreased, and the filling of the packing was continued using a new value of r_c. This procedure gives us a set of configurations filled in for the given values of r_c.

The composition of dense polydisperse packings are usually studied using the "weight" distribution function $m(r) = \frac{4}{3}\pi r^3 n(r)$, where $n(r)$ is the particle size distribution. Figure 2 gives $m(r)$ for two models filled up to cutoff length $r_c = 0.05$ and having $\eta_1 = 0.856$ and $\eta_2 = 0.862$. The initial configuration for the first model had $\eta_1^0 = 0.300$ and was constructed by the random throwing of hard spheres into the model cube. The initial configuration for the second model had $\eta_2^0 = 0.550$ and had been obtained by compressing the first initial configuration by the Monte Carlo method. The particle size distribution of the initial configurations was given by the Gaussian law with a mean value $\bar{R} = 0.5$ and standard deviation $\sigma = 0.05$. The statistical noise in the distribution in the region of "coarse" particles (most of them are the initial ones) was decreased by using larger initial configurations containing 2000 particles. However, due to computer limitations we could fill in such packing up to a cutoff length of about $r_c = 0.2$. The region of "fine" particles was reached starting with the initial configurations of 100 particles. The curves in Fig. 2 have been obtained by linking the corresponding parts of the distribution at $r_c = 0.2$.

A detectable difference in $m(r)$ of our models in the region of coarse and mean-sized particles $(r > 0.3)$
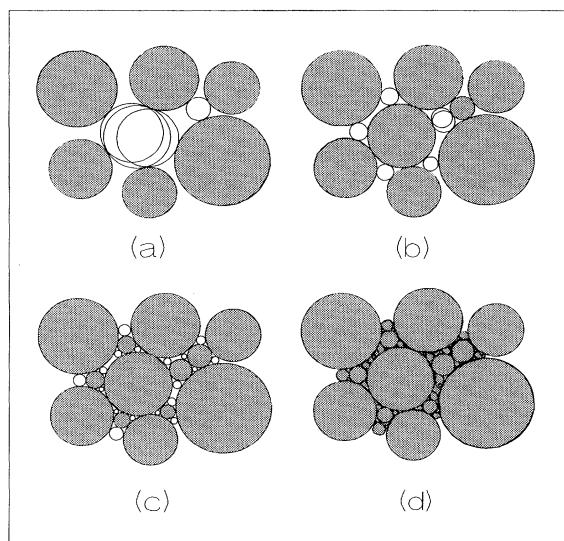

FIG. 1. A two-dimensional illustration of the packing procedure. All interstitial spheres (empty circles) are determined for a given initial configuration of spherical particles (full disks) (a). Some of the interstitial spheres are turned into new particles of the system (the largest sphere is chosen from the overlapping ones). In a result, we have a new configuration of particles and calculate a new set of interstitial spheres (b). In a similar way, nonoverlapping interstitial spheres are turned into new particles of the system (c). Repeating the procedure many times, one can fill in the space with any high density (d).
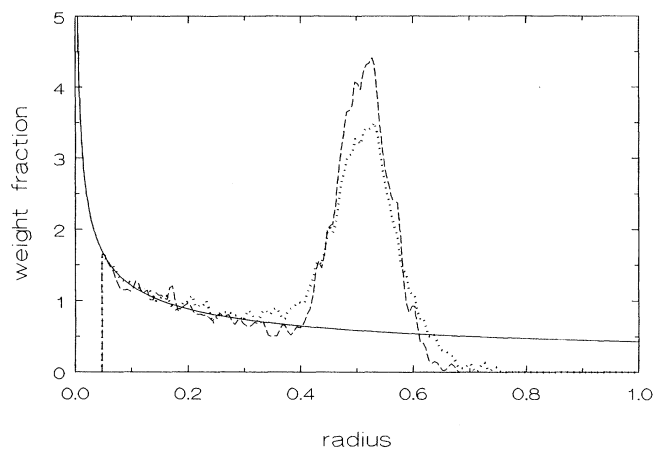

FIG. 2. Weight fraction of particles in the packings vs particle radius. Dotted and dashed lines correspond to the models obtained from initial configurations of different density (see text). Solid lines are calculated from the formula $m(r) \sim r^{-0.45}$, which describes the asymptotic behavior for the Apollonian packings within "fine" r. The packing fractions of the models are $\eta_1 = 0.865$ and $\eta_2 = 0.867$.

depends on the initial configurations. On the other hand, one can see that the weight composition of fine particles ($r < 0.3$) is the same for both models. Additional calculations showed that the composition of fine particles is also insensitive to the form of the initial particle size distribution. Moreover, $m(r)$ in this region is well described by the asymptotic law $m(r) \sim r^{-D+2}$ with $D = 2.45$ (see below).

This weight distribution can be compared with experimental results. The available bimodality of distribution as well as the relation between the midsized and coarse particles are in accord with the conclusions of the experimental paper [11], where the polydisperse packings of glass spheres of the same density were obtained. Besides, our calculations are pointing to the universal behavior of the "fine particle." However, special investigations are necessary to make real packings with the Apollonian particle size distribution.

We have also studied the fractal properties of our models. It is known that the Apollonian packings are self-similar [12]. However, so far only two-dimensional systems have been studied [21–23]. In such fractals the number of particles $N(r_c)$, the radii of which exceed a given cutoff value r_c, obeys the law $N(r_c) \sim r_c^{-D}$, where D is the fractal dimension. Our computer models can readily be employed to determine the D value by drawing the $N(r_c)$ dependence in the logarithmic coordinates (see Fig. 3). For small r_c the dependence could be well approximated by a straight line whose slope yields the D value of 2.40. The fractal dimension can also be estimated from the porosity [22]. The relation $\varepsilon(r_c) \sim r_c^{-D+3}$ holds for three-dimensional fractals. The corresponding dependencies for our models are also given in Fig. 3. The fractal dimension estimated from the porosity is 2.50. The coincidence of this value with that estimated from the number of particles $N(r_c)$ is considered to be satisfactory, taking into account a relatively small change of the length scale in our models (the particle radii differ a bit more than by a factor of 10). We estimate the value of D as 2.45. This value is obtained equally for models created from initial configurations of different density (see Fig. 3). Our additional calculations show that more broad initial radius distribution ($\sigma = 0.15$) gives the same result. Nevertheless, the fractal dimension of the packing may depend on the method of preparation, e.g., if the interstitial spheres with smaller radii are preferred to larger ones. This is in contrast to our procedure. For two-dimensional Apollonian packings the different authors give D values from 1.3 to 1.5 [21,22].

We have noticed when creating the models that the mean number of faces of the Voronoi S regions $\bar{f}$ decreases monotonously with the number of iterative steps of the procedure. For initial configurations and after several first steps of the procedure, we obtain $\bar{f} = 14.5$, which is close to the value of the mean number of the faces of the Voronoi polyhedra in the disordered packings
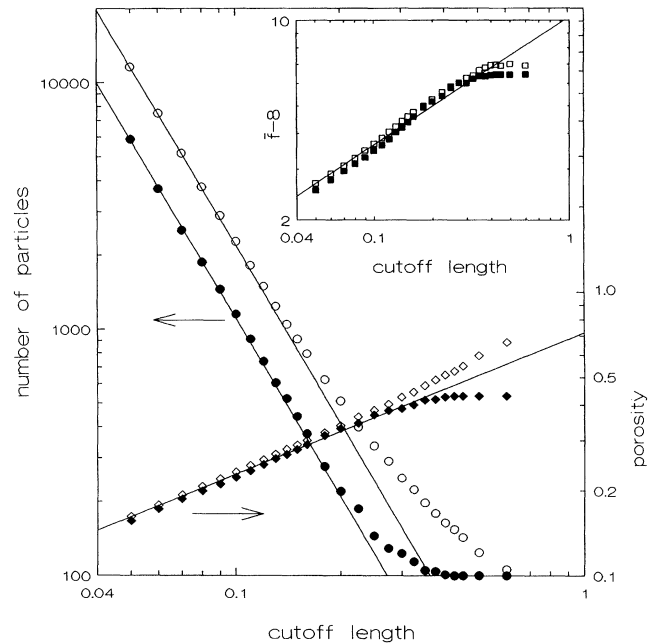

FIG. 3. The log-log dependence of the number of particles $N(r_c)$ (circles) and porosity $\varepsilon(r_c)$ (rhombs) for our packings vs the cutoff length r_c. Open symbols refer to the model constructed from the initial configuration of the low density $\eta_1^0 = 0.300$; closed symbols are from $\eta_2^0 = 0.550$. The same dependences for the average number of the faces of the Voronoi S regions $\bar{f}$ are in the inset.

of equal spheres [14]. At the end of our calculations (after a few tens of steps) the $\bar{f}$ falls to about 10. The reason is clear from a two-dimensional illustration in Fig. 4. A new particle inscribed into the system at a far step of the procedure has, as a rule, only three geometrical neighbors; i.e., the Voronoi S region of the given particle has three faces. As a result, the topology of the Voronoi network will be similar to that depicted in the inset of Fig. 4. This pattern resembles the Sierpinski carpet [12]. For a three-dimensional case, the new particles are sure to have only four geometrical neighbors, and the Voronoi network is similar to the three-dimensional Sierpinski carpet. From this, one can calculate that the $\bar{f}$ value converges to 8 for a three-dimensional Apollonian packing.

We have calculated the averaged number of faces of the Voronoi S regions $\bar{f}$ for our models as a function of the cutoff length r_c; i.e., the Voronoi S regions were constructed for configurations filled in up to a given value of r_c. The inset of Fig. 3 demonstrates the dependence of $\bar{f} - 8$ on r_c in the logarithmic scale. For small r_c, it can be approximated by a linear function with the tangent of the slope angle being 0.45. Note, this value is close to that for the porosity (see also Fig. 3).

In summary, we are proposing a method to calculate three-dimensional Apollonian packings. It has been used to create computer models of dense packings of polydis-
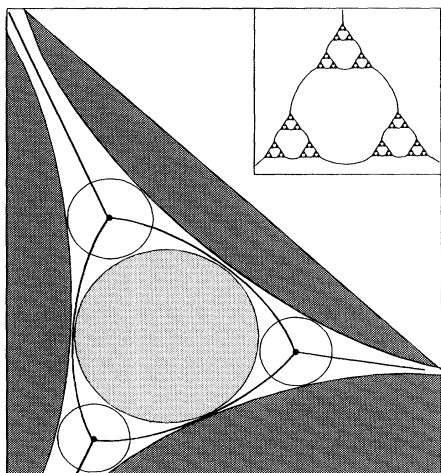
FIG. 4. A two-dimensional illustration of inscribing a new particle into the model at the far iterative step of the procedure. A new particle has only three geometrical neighbors: three faces of the Voronoi S region. (For three dimensions it has four faces.) The Voronoi network of the model is (asymptotically) similar to the one depicted in the inset.

perse hard spheres. Having at hand such models one can predict particle size distribution to get a packing with high bulk density [11]. The fractal nature of such models is investigated. This property of dense polydisperse packings should be taken into account in the interpretation of the fractal behavior obtained for the "Bernal-type" models [24,25]. One should also have in mind the fractal nature of the Voronoi network of such packings. It is interesting for percolation and diffusion problems, because the Voronoi network is a "navigation map" of the interparticle volume. Flows and impurity particles move along the bonds of the Voronoi network [13,17].

[1] C. C. Furnas, Bur. Mines. Rep. Invest. **2894**, 7 (1928).
[2] J. Bernal, Proc. R. Soc. London **280**, 299 (1964).
[3] J. E. Ayer and F. E. Soppet, J. Am. Chem. Soc. **48**, 180 (1965).
[4] W. S. Jodrey and E. M. Tory, Phys. Rev. A **32**, 2347 (1985).
[5] N. Standish, A. B. Yu, and R. P. Zou, Powder Technology **68**, 175 (1991).
[6] A. B. Yu and N. Standish, Powder Technology **52**, 233 (1987).
[7] N. Ouchiyama and T. Tanaka, Ind. Eng. Chem. Res. **28**, 1530 (1989).
[8] R. Jullien, P. Meakin, and A. Pavlovich, in *Growth of Packings in Disorder and Granular Media,* edited by D. Bideau and A. Hansen (Elsevier Science Publishers B.V., New York, 1993), p. 103.
[9] W. Soppe, Powder Technology **62**, 189 (1990).
[10] Y. Konakawa and K. Ishizaki, Powder Technology **63**, 241 (1990).
[11] D. C. C. Lam and M. Nakagawa, J. Ceram. Soc. Jpn. **102**, 133 (1994).
[12] B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman and Company, New York, 1983).
[13] N. N. Medvedev, Dokl. Akad. Nauk **337**, 767 (1994) [Dokl. Phys. Chem. **337**, 157 (1994)].
[14] J. L. Finney, Proc. R. Soc. London A **319**, 479 (1970); **319**, 495 (1970).
[15] D. Rigby and R. J. Roe, Macromolecules **23**, 5312 (1990).
[16] S. K. Chan and K. M. Ng, Powder Technology **54**, 147 (1988).
[17] S. Bryant and M. Blunt, Phys. Rev. A **46**, 2004 (1992).
[18] M. Tanemura, T. Ogava, and N. Ogita, J. Comput. Phys. **51**, 191 (1983).
[19] N. N. Medvedev, J. Comput. Phys. **67**, 223 (1986).
[20] B. J. Gellatly and J. L. Finney, J. Non-Cryst. Solids **50**, 313 (1982).
[21] D. W. Boyd, Mathematica **20**, 170 (1973).
[22] H. J. Herrmann, G. Mantica, and D. Bessis, Phys. Rev. Lett. **65**, 3223 (1990).
[23] S. S. Manna and T. Vicsek, J. Stat. Phys. **64**, 525 (1991).
[24] P. Pfeifer, D. Avnir, and D. Farin, in *Large-Scale Molecular Systems,* edited by W. Ganes et al. (Plenum Press, New York, 1991).
[25] P. Levitz, J. M. Drake, and J. Klafter, J. Chem. Phys. **89**, 5224 (1988).