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Coordination number of disordered packings of identical spheres

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Abstract. We determine the coordination number in ordered and disordered packings of monosize spheres, following a first treatment given by Elizabeth Gardner. The average number of contacts per unit volume is derived from the slope at the origin of the distribution law for the separators obtained by passing a random line across the assembly of spheres. Moreover, this law may be completely calculated and is universal up to a multiplicative constant. We have performed tests on numerical ordered and disordered assemblies of spheres and obtained the complete distribution function for the separators. We discuss the difference between the theoretical-biparticle and stereological approaches and discuss how the study of the pair correlation function analysis may extend the present treatment.

1. Introduction

Most transport properties of granular media depend on the geometry. The application of stereology based on the study of random cuts of such media provides statistical information on the packing such as its porosity, interfacial area per unit volume and so on, provided certain properties such as isotropy and homogeneity are met [1]. Unfortunately, no similar result exists in general for the coordination number Z, i.e. the average number per grain of real contacts between grains which plays a crucial role in the transmission of current forces in those arrays [2]. In addition, the definition and distinction between actual and near contacts is a delicate problem [3].

Packings of spheres provide rather convenient models for the study of real or theoretical granular media and their geometry is particularly well documented [4]. It is therefore a privileged system for seeing how the coordination number can be extracted from random cuts by planes (2D) or lines (1D). However, even in the case of packings made of monosize spheres, where the geometry of the grain is simple, theoretical predictions as well as experimental results on Z vary according to the particular determination used [5]. A few years ago, Pomeau and Serra [6] derived the average number n_c of real contacts per unit volume from the limit behaviour of the distribution function of the closest distances between the disc cuts obtained in a plane section. Chermant *et al* [7] have developed an experimental program on the basis of this approach and have applied it to regular dense packings where the theoretical value of n_c is known exactly.

More recently, Elizabeth Gardner, in a short letter to Journal de Physique [8], proposed that the information given by a random line drawn through the medium

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would provide a simpler answer to the coordination number problem than the previous solution of Pomeau and Serra. Her method involved the distribution function of the distance between two consecutive intercepts of spheres in contact that we will call a *true separator* in the following. She also extended her analysis to a general distribution of polydisperse spheres. She finally proposed that the analysis might be extended to more general classes of objects.

We have recently revisited Gardner's calculation by a different method and, apart from a numerical factor, recovered her result [9]. We have also calculated the distribution function for true separators, which is a universal function up to a multiplicative constant which is merely equal to $n_c R^2$ and which can be derived from the slope of the curve at the origin. We have also validated her approach on numerical packings of spheres of equal radius. Further extensions involving the radial correlation function to non-touching particles can be investigated; we shall consider briefly this aspect only in the last section.

This paper is devoted to the numerical part of our study. Theoretical calculations may be found in our previous paper [9] and will be omitted. In § 2, we recall the Pomeau and Serra approach and present our analysis following that of Gardner. In § 3, we extend our presentation of the numerical study of ordered and disordered packings of spheres and check the universality of the distribution function. The difference between the theoretical and stereological analysis is stressed. In the last section, we compare the 1D and 2D approaches and we discuss openings of Gardner's work.

2. Theoretical approaches

Let us first recall here the 2D stereological approach to the coordination number given by Pomeau and Serra before presenting our approach following Gardner's original idea. A comparison will be made in the last section. The complete calculation of the distribution function for the true separators may be found in our previous paper [9].

2.1. Planar sections

In planar sections, assemblies of spheres in contact are represented by assemblies of non-touching discs of various radii whose centres are the projections of the centres of the spheres and whose radii r_i are the projected lengths of the radii R of spheres i (figure 1); the line which joins the centres of the projections of two discs defines the smallest distance ρ_{ij} between the discs (figure 2(a)).

The calculation of the average number of real contacts relies on the statement that when the minimum distance between two circular sections is small, it corresponds to two spheres in contact; this is rigorously true for ordered packings, as the distance between spheres takes fixed values and that, below a minimum gap value, two spheres have to be in contact. It is only a limit law for disordered packings as the distance between spheres varies continuously.

For monosize spheres of radius R, the average number per unit area of neighbouring discs with closest distance smaller than ρ is given by

$$F(\rho) \simeq n_c \sqrt{\pi \rho R}\, \frac{\Gamma(7/4)}{\Gamma(9/4)} \tag{1}$$

for small ρ. It is then relatively easy to get function $F(\rho)$ for any ρ through image analysis and to extrapolate it at the origin. The coordination number Z is related to

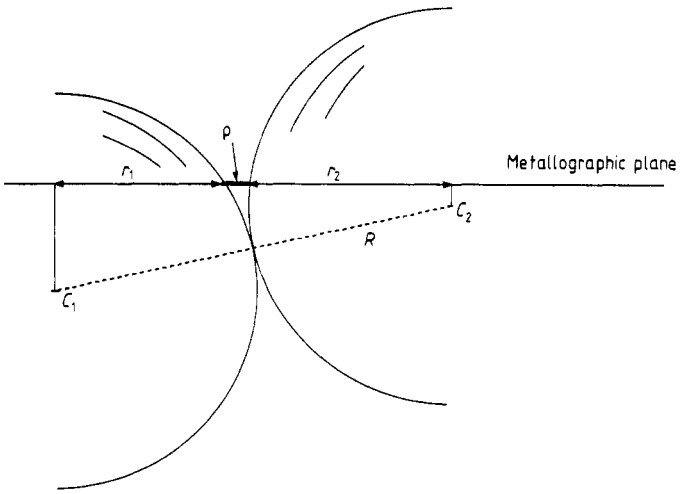


Figure 1. View of a 2D cut of a biparticle of two spheres.

n_c through

$$\frac{Z}{2}=\frac{n_c}{n_v} \tag{2}$$

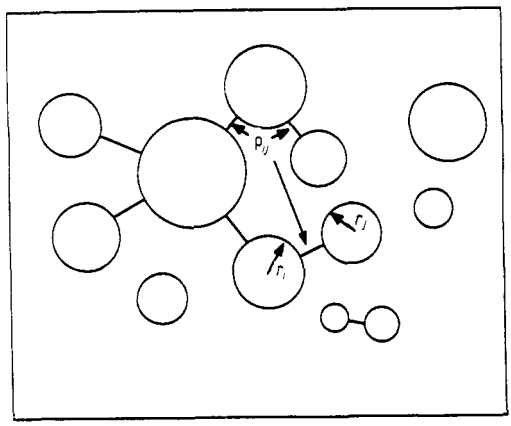
where n_v is the average number of spheres per unit volume. Formula (1) has been checked experimentally on a face-centred cubic array [7] and has given, with good accuracy, the expected value $n_c=3/(2\sqrt{2}R^3)$. The complete function $F(\rho)$ is an increasing function of ρ which saturates for large ρ. Its analytic expression is not known (the Pomeau and Serra calculation explicitly stated that the distance ρ is small) though some results seem to have been implicitly assumed by Chermant *et al* [7].

Actually, when $\rho \geq 2R(\sqrt{2}-1)$, the count given by $F(\rho)$ includes second-nearest neighbours as it is not possible to separate sections coming from touching and non-touching spheres with image analysis. One may ask whether other information may be obtained from $F(\rho)$, especially for non-touching spheres. It is expected that the radial (or the pair) distribution function then plays an important role [10].

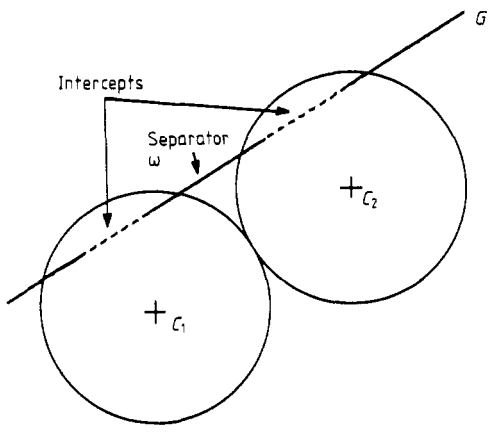
2.2. Line intersections

The idea of this treatment is to consider the intersection of a random line with the spheres of the packing. A line which crosses one sphere determines, within the sphere, a chord which is called an 'intercept'. The distance between two intercepts on the line is called a 'separator' (figure 2(*b*)); when spheres are in contact, the separator is called a 'true separator'.

It is possible to get the number of separators per unit line smaller than a given value ω by the same kind of analysis as above. Let $N_L(\omega)$ (L stands for line) denote the corresponding function. For small ω, it coincides with the number $N_L^*(\omega)$ of true separators smaller than ω as, again, small separators arise from touching spheres. We have obviously $N_L(\omega) \geq N_L^*(\omega)$. However, unlike the situation of § 2.1, the complete calculation of $N_L^*(\omega)$ has been performed [9]. A main feature is its universality: the reduced function $N_L^*(u)/(n_c R^2)$ is found to be a unique function of the reduced variable $u=\omega/2R$. It is plotted in figure 3 for $0<u<1$ (full curve).



(*a*)



(*b*)

Figure 2. (*a*) Measurement of ρ in a planar section; (*b*) Intersection of a biparticle by a line.

The behaviour of $N_L^*(u)$ is, in the limit where u goes to zero:

$$N_L^*(u) \simeq \frac{2\pi}{3} n_c R^2 u. \tag{3}$$

n_c and, consequently, the coordination number Z can be obtained from the slope at the origin of this law. In the other limit $u = 1$, $N_L^*(u)$ saturates to its maximum value as the largest true separator is $2R$.

If a dense packing of monodisperse hard spheres is regarded as a packing of biparticles in contact with a volume density equal to n_c we can calculate the value $N_L^*(u = 1)$ from the geometrical problem of a single biparticle crossed by random lines. We have found

$$N_L^*(u = 1) = \pi n_c R^2 \left(1 - \frac{8}{3\pi}\right). \tag{4}$$

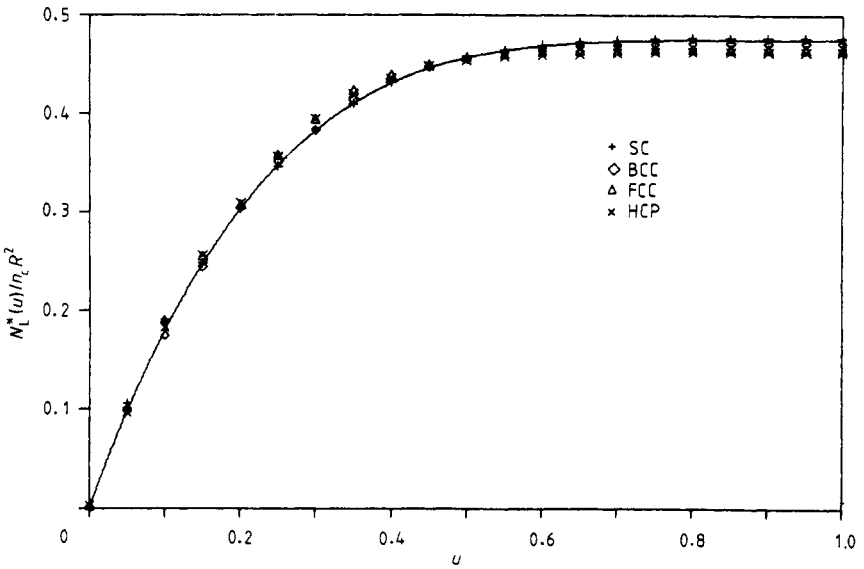


Figure 3. Plot of the reduced function $N_L^*(u)/(R^2 n_c)$ against u. The full curve represents the theoretical curve and the four kinds of symbols the normalised distribution functions for the four ordered 3D packings (SC, BCC, FCC and HCP).

Unfortunately, the above hypothesis is not directly applicable to a stereological study (see § 3.3), and the experimental values obtained on any packings (ordered or not) will be lower than those given by relation (4).

As a by-product, let us recall the following classical stereology result. The number of separators per unit length is related to n_v, the number of spheres per unit volume, by $N_L = \pi n_v R^2$. But the ratio

$$\frac{N_L^*(u=1)}{N_L}$$

must be smaller than 1. We deduce an upper bound of the coordination number

$$Z_{max} = \frac{2}{[(1-(8/3\pi)]} = 13.23 \tag{5}$$

which is not far from the values 13.56 and 13.40 obtained by Coxeter [11] and Dodds [12].

3. Numerical simulations

We checked the validity of our calculations on simulated ordered (SC, BCC, FCC or HCP) and disordered packings of equal spheres. Technical details are given in [9].

3.1. Ordered packings

We have performed numerical simulations on finite cubic samples with size from $10 \times 10 \times 10$ up to $40 \times 40 \times 40$ and a number of lines from 20 up to 600 to estimate the

range of values needed to obtain a good accuracy of the slope of $N_L(u)$. We present results on $20 \times 20 \times 20$ numerical packings. In order to compensate for this limited size and for the effect of ordering of the packings and to obtain smoothly shaped distribution functions, 300 lines were thrown through the packings. The slope at the origin was calculated by a linear regression with the constraint that the resulting line passes through the origin. This calculation was carried on with values of u up to 0.04. For each type of ordered packing, ten simulations were computed in order to average the ten corresponding slopes. The values of n_c derived from these mean values coincide with the known exact values with an error lower than 4%.

As numerical packings enable us to check whether two spheres are in contact or not, we may obtain the full normalised function $N_L^*(u)/(n_c R^2)$. The resulting function is to be compared with the theoretical one [9] (see figure 3). The agreement is excellent up to an upper limit on u which will be given in § 3.3. It does not depend on the lattice under consideration. As noticed in § 2.2, systematic differences between the experimental curves and the theoretical one must be again pointed out and are explained in § 3.3.

3.2. Disordered packings

We used numerical 3D packings constructed with the Powell algorithm [13]. We created several packings (from 1000 up to 3000 spheres) inside cubes varying from 10 to 15 times the sphere diameter. The packing fraction fluctuates between 0.590 and 0.605 which can be compared with the real packing fraction of a monosize packing made of glass beads (around 0.62). We have thrown 300 lines through these packings to get a satisfactory statistical account.

From this numerical experiment we get a value of n_c equal to 0.427 ± 0.025 which is to be compared with the exact one 0.429 obtained from the previously calculated density; the mean coordination number, given by (2) is $Z \simeq 6.1 \pm 0.3$. Universality of the function $N_L^*(u)$ may again be checked starting from the normalised function.

We have plotted on figure 4 the complete function $N_L(u)$ for all separators together with $N_L^*(u)$; we see directly that they have a common slope at the origin, which justifies *a posteriori* the method for determining n_c. They separate rapidly and $N_L(u)$ keeps growing while $N_L^*(u)$ reaches a limit value much sooner.

Let us point out that it is not possible to distinguish experimentally on a planar or a linear section between distances corresponding to touching spheres and those which are not in contact. Thus, we cannot analyse separately at the same time contact and non-contact functions such as $N_L^*(u)$ and $N_L(u)$ in the present case (or $F^*(\rho)$ and $F(\rho)$ in the previous analysis). Practically, only $N_L(u)$ and $F(\rho)$ are relevant in a stereological experiment.

3.3. Theoretical and stereological functions

It is important to emphasise the difference between the theoretical and the experimental approach.

The theoretical calculation, as stated above, considers only biparticles, independently of other surrounding spheres. Then, any pair of spheres is considered and the distribution law for u is given only by the probability of a line being a separator for the biparticle.

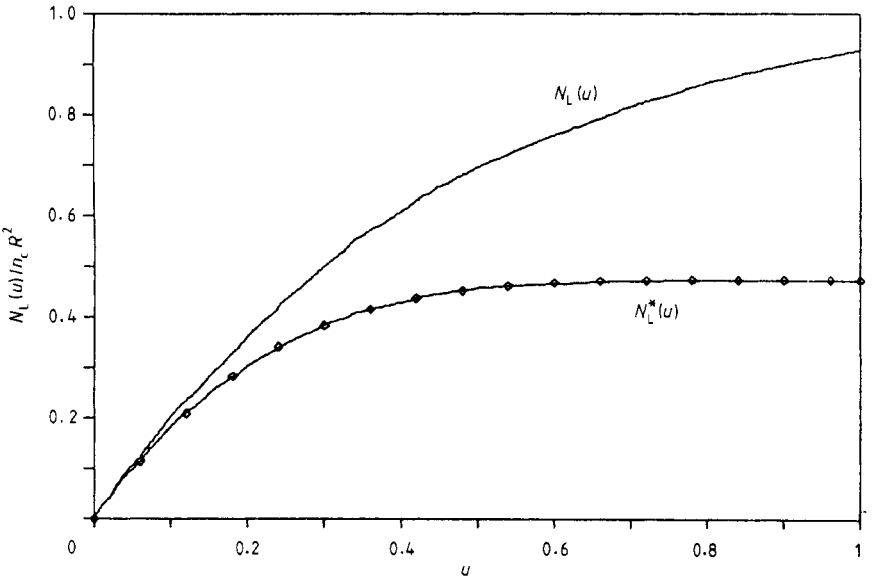


Figure 4. Distributions $N_L^*(u)$ and $N_L(u)$ obtained from disordered packings of monosize spheres. The curves have the same slope at the origin, equal to 0.427 ± 0.025.

The stereological approach takes into account the environment of a biparticle. It strives to describe the pore space as it considers only separators without any interference with a third particle. A fraction of the configurations which are considered in the theoretical calculation are then omitted as seen in the limit configuration shown in figure 5. Thus, we have $N_L(u)^{\text{stereo}} \leqslant N_L(u)^{\text{theor}}$.

The two values coincide only for $u < u_0 = (1 - \sqrt{2\sqrt{3} - 3}) = 0.318$; beyond this value, they separate. The discrepancy is very important in the long-range limit as the counting in $N_L(u)^{\text{stereo}}$ slows down for large u: two spheres which are far apart are nearly always separated by a third one. Similarly, for the contact functions, we have $N_L^*(u)^{\text{stereo}} \leqslant N_L^*(u)^{\text{theor}}$.

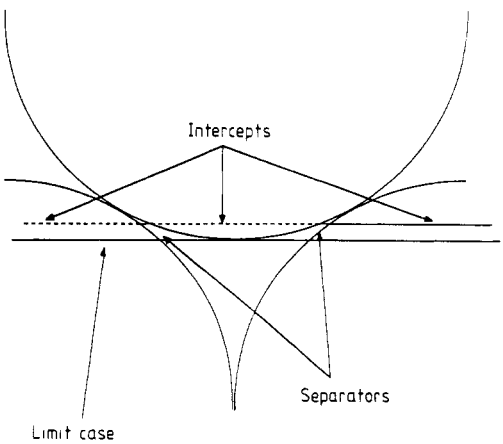


Figure 5. Limit case where stereological and theoretical $N_L(u)$ coincide. The linear cut is in the plane of the three centres and is tangent to the upper circular section.

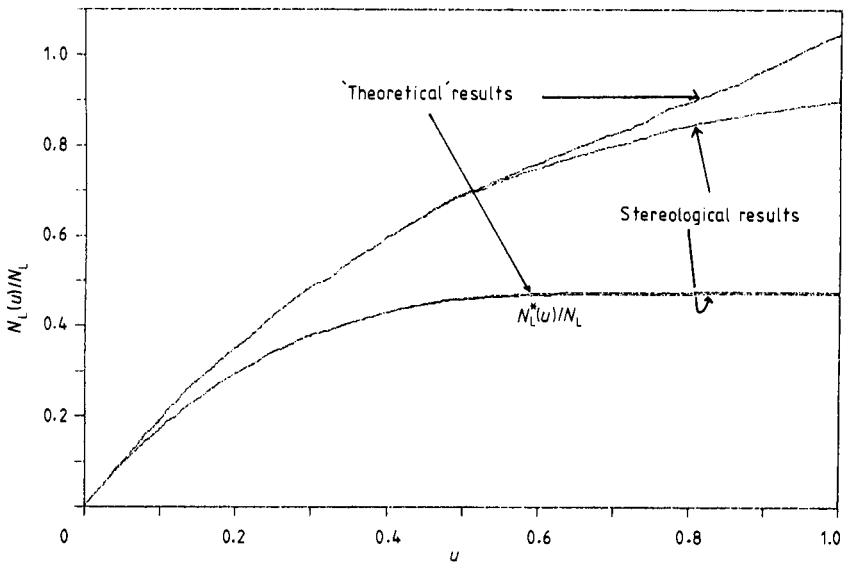


Figure 6. Plot of the four reduced distribution functions on the line $N_L^*(u)/N_L$ and $N_L(u)/N_L$ obtained 'theoretically' and stereologically.

We have plotted in figure 6 the four distribution functions related to the line analysis in the case of a numerical disordered packing [14]. $N_L^*(u)^{theor}$ and $N_L^*(u)^{stereo}$ are not very different as configurations where true separators are omitted are relatively rare. On the other hand, $N_L(u)^{theor}$ and $N_L(u)^{stereo}$ very rapidly differ as expected (in the case of regular packings, the differences between $N_L(u)^{theor}$ and $N_L(u)^{stereo}$ give the relative position of the second and third neighbours).

4. Discussion

We have presented in this paper a 1D method for estimating the coordination number of a monosize packing of spheres from the limit behaviour of the distribution functions $N_L^*(u)$ or $N_L(u)$ per unit line as the length of the separator goes to zero. The distribution function $N_L^*(u)$ can be written explicitly, provided homogeneity and isotropy conditions are fulfilled. It is universal and its expression has been tested both on ordered and disordered numerical packings. The difference between the biparticle and the stereological approaches has been emphasised. The discussion may be extended to binary mixtures, to assemblies of spheres with size distribution, and to 'soft' contacts, which is a first step towards slightly sintered grains. This will be done in the last section. A more complete presentation will be given in a forthcoming paper [14].

4.1. Possible experiments

The method proposed by Gardner does not look technically very far from that of Pomeau and Serra as both analyses can be made on a random cut of the packing. One difference is that, in the case of a linear analysis, the full function $N_L^*(u)$ is known; we do not have a corresponding calculation for $F^*(\rho)$ which is the part of the function

$F(\rho)$ which only retains the values of ρ when the spheres are in contact. There is another large difference which comes in the treatment of experimental packings: in the Pomeau-Serra approach, several cuts are needed in order to obtain a large enough number of couples of discs in order to minimise the error on the slope of $F(\rho)$ [7]; in Gardner's approach we use more fully the information on the relative position of one sphere with respect to the others by throwing a large number of lines, even in a single 2D cut [14].

An experimental study of the problem has been undertaken and will be presented separately. Let us mention the condition of the experiment. We create packings of spheres with well known procedures which lead to a global homogeneity. Then, sections are obtained in the following way: the pore space is filled first under vacuum with a low viscosity coloured epoxy resin in order to obtain a compact solid block. Plane random cuts are made through the block and polished. They are studied by image analysis, pixel after pixel (512×512 pixels are necessary). In more recent experiments carried out with J L Bouillot, we have scanned the surface by parallel equidistant lines using an image analysis technique which determine the precise location of the points where the light intensity changes. We use an algorithm initially developed [15] for the study of spheres located in a plane. The treatment leads to the reconstruction of the centre of the discs (or spheres in his case) from the intercept of at least three lines intercepting a given circle in a plane. An example of such a treatment is given in figure 7. Moreover, preliminary results on numerical packings have shown us that the 1D study could also be performed by generating random lines within a large enough 2D cut only, or using parallel *non-coplanar* lines, with directions chosen at random, and distant by more than $R\sqrt{2}$ in order to decorrelate the information between two neighbouring lines.

This method of cutting is rather long and destructive. Moreover it is not a direct experimental 1D study. One can imagine an experiment with a laser beam passing through a three dimensional packing of beads filled with a fluorescent isoindex liquid as has been used for the study of multiple phase flows in porous media [16]. This will give directly without damaging the packing, the position of the respective intercepts of the packing with the line and, therefore, the distribution law of the separators.

4.2. Extensions of Gardner's work

In the spirit of the last section of Elizabeth Gardner's letter, we consider some possible extensions beyond monodisperse arrays of spheres.

4.2.1. Binary disperse case. Gardner's treatment led to the full distribution of contacts in polydisperse arrays of objects in contact. We consider the simpler limit of a bidisperse random array of spheres of radii R_1 and R_2 ($R_1 < R_2$). The study of such arrays is a well documented subject [17]: packings of binary spheres are often used as a model for more general-continuum distributions of spheres whose radii R verify the relation $R_1 < R < R_2$. On the theoretical side, several approaches have been developed for the study of random binary alloys, such as the Percus-Yevick method. Geometrical treatments also exist: they lead to the determination of the packing fraction of spheres as well as to predictions concerning the fractions of contacts between spheres 1 and 2 (respectively t_{11}, t_{12} and t_{22} such that $t_{11} + t_{12} + t_{22} = 1$) [12]. On the experimental side, we have developed a technique which provides reasonably homogeneous and

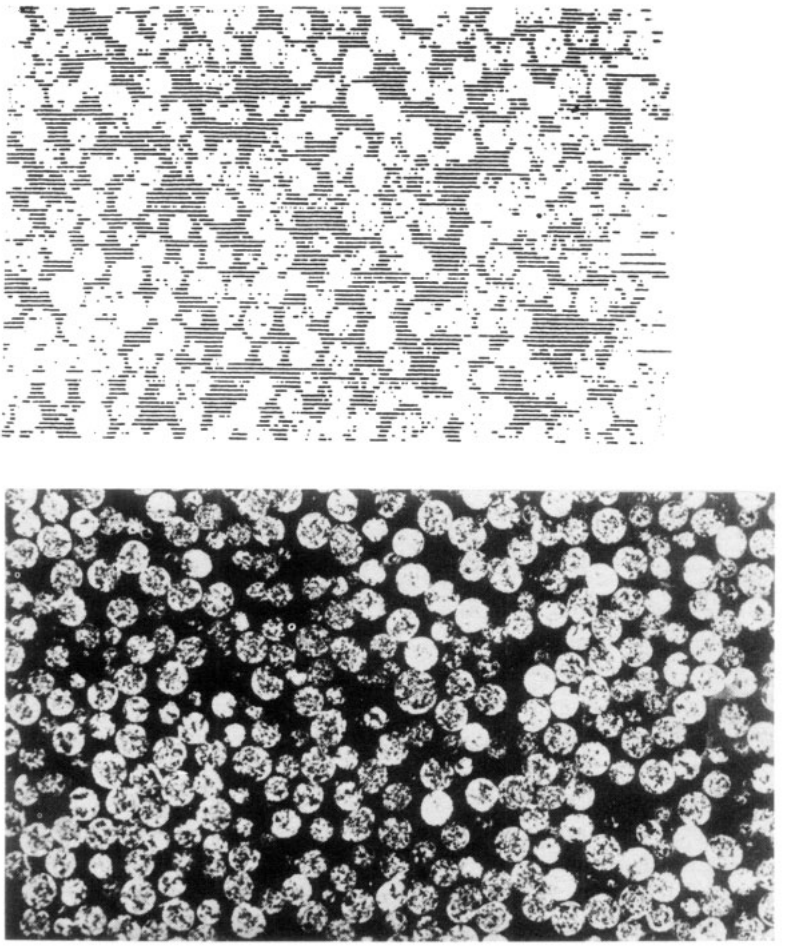


Figure 7. Photography and analysis of a 2D cut of a spheres packing made by the technique developed in Bouillot's thesis.

isotropic packings of spheres provided the ratio R_2/R_1 is sufficiently small to avoid macroscopic segregation of spheres (typically smaller than 4) [18].

If we apply Gardner's treatment to such distributions, we find

$$N_{\mathrm{L}}^{*}(\omega) \simeq \frac{\pi}{3} n_{\mathrm{c}} \omega \left(R_1 t_{11} + \frac{2R_1 R_2 t_{12}}{R_1 + R_2} + R_2 t_{22} \right).$$

We recover the expected result $(\pi/3) n_{\mathrm{c}} R \omega$, in the limit $R_1 = R_2$.

4.2.2. Sintered monosize sphere packings. Another opening deals with overlapping spheres. Such a situation is met in several instances in materials science.

Spheres having a hard core and soft shell are used to model microemulsions [19].

Toughness studies of polymers reinforced with hard spheres show that the presence of mechanical contacts between spheres is not needed to insure a continuous rigid path but that there exists a critical radius (larger than the sphere radius) where the continuum medium will resist shearing [20].

Overlapping spheres are also used to model the sintering of spheres [21]. One starts from compact arrays and then dilates the spheres without changing the location of their centres. The excess material in the overlapping regions is not taken into account. This description is not too far from reality as studied on slightly sintered glass bead packings (figure 8).

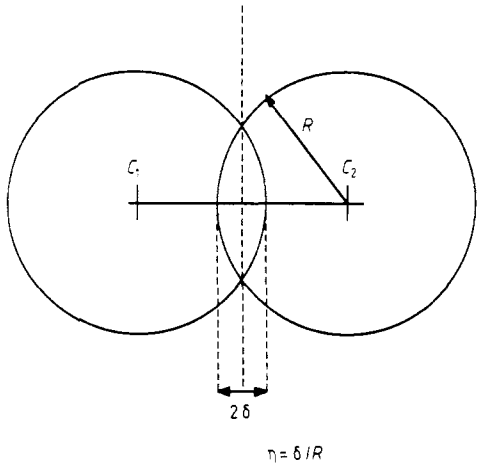


Figure 8. View of a 'theoretical' sintering process.

Pomeau and Serra [6] have considered the extension of their calculation to this case. It leads in particular to a less singular distribution of minimum lines near a zero separation. We have also extended Gardner's calculation to this case and found the following formula [14]:

$$N_L^*(u) = \frac{2\pi}{3} n_c \frac{1-4\eta+2\eta^2}{1-\eta} R^2 u \qquad \text{for small } \eta$$

with $\eta = \delta/R$, δ representing the overlapping part of one sphere.

This leads indeed to the formula we obtained in the case of monodisperse packing of hard spheres when $\eta = 0$:

$$N_L^*(u) = \frac{2\pi}{3} n_c R^2 u.$$

4.2.3. Other possible extensions. It remains to see what can be done beyond the properties of contact to describe the immediate environment of spheres, i.e. what can be extracted from the knowledge of the complete distribution function. A similar problem has formally been solved starting from the 'intercentres' of the sections. However, an unresolved problem remains which may be raised in some cases [10]. In a first step, it seems easier to consider the biparticle approach, as all pairs may be taken easily into account in the image analysis procedure by suitable changes of the studied area. It is relatively easy to extend the calculation done in the case of touching spheres to all kinds of biparticles. One main objective which we are currently pursuing is to determine $g_v(r)$, the two-point correlation function in 3D space, by inverting—at least numerically—the 1D-3D equation.

The final paragraph of Elizabeth Gardner's letter is the most provoking one. It raises the possibility that her approach might be used to describe the distribution of contacts for general packings which was the starting point of the present paper. To the best of our knowledge there has been no stereological work done on this subject. However in the spirit of the current issue we would like to propose some paths to extend Elizabeth Gardner's thoughts. Can we think of using some particular geometries of objects which would best mimic general random packings?

Obviously, the polydispersity in the distribution of spheres provides an insufficient step and merely generalises the monodisperse results.

Ellipsoids of known geometries could be used in some limit cases (small or large ellipticity) instead of spheres to mimic randomly shaped grains. In such a case, it will be easy to come back from the 2D cuts to a 3D reconstruction and one can think of a brute-force solution for calculating the number of contacts. The problem is likely to be overdetermined as in case of spheres [22]. However this approach would take us away from the original approach of Elizabeth Gardner. In addition, problems of packings of ellipsoids (at the limit cylinders) are far more complex than those of spheres mainly because of the orientational degrees of freedom.

It may turn out that the simplest system to deal with would be a distribution of regular, randomly shaped convex objects (potato type) with a given range of size. It is well known from integral geometry that the problems of coating and local environment of objects are much simpler if such a convexity property applies, which is often the case in nature. Thanks to this randomness we would not have orientational order. The interest of such a distribution is that a random cut by a plane or a line of two touching objects will be characterised by smooth convex curves which—as an average over many objects—can mimic the planar circular arc next to the point of minimum separation in the cut of a sphere. It would be tempting to apply directly the Gardner or Pomeau-Serra treatments to such assemblies and, on the other hand, to check the real number of contacts of the three-dimensional objects. This can be done by several techniques which have been developed for this purpose such as looking for print points left on the contacts due to the applied pressure, or a deposit left when a solution which has stayed in the medium for a while and washed away.

As stated early in this paper, the practical importance of such a study is to achieve a better grasp of the transport properties in electrical [2] as well as mechanical [3] granular arrays. Another problem is the formation of ceramics or sintered materials in which the initial steps in the process (formation of a paste or packing) play a dominant role for the final material. Some theoretical studies [23] have recently considered this problem. These approaches clearly indicate that the heterogeneity in the location as well as in the quality of contacts is an important factor to be taken into account which is a problem beyond the present statistical evaluation.

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