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# 1D stereological cuts : application to slightly sintered model materials

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**Résumé.** — La mesure des intercepts engendrés dans des empilements granulaires par des coupes linéaires aléatoires peut fournir plus simplement que des sections planes certaines informations concernant la géométrie de ces milieux. La méthode, déjà testée sur des empilements de grains sphériques durs est ici utilisée pour déterminer le nombre moyen de « cous » par grain dans un modèle classique de matériau légèrement fritté.

Abstract. — Intercepts generated through granular packings by random 1D (line) sections may provide information on the geometry of the medium in a simpler way than metallographic cuts. The method, already tested on hard monosize spherical grain stackings, is used here to determine the average number of necks per grain of a classical model for slightly sintered materials.

#### 1. Introduction.

Sintered materials play a more and more important role in everyday life (ceramics, glasses, optical fibers, powders...) and are now being extensively studied. Part of their physical properties depend on their geometry, a good tool for investigation seems to be stereology [1] combined with image analysis [2]: some metric parameters such as porosity are directly derived from the quantitative analysis of planar sections. With complementary assumptions about the structure, we can obtain other 3D parameters such as the mean size of the cavities or coordination number. They are related to physical properties [3, 4] in a semi phenomenological way. For example, conductivity in the pore space is proportional to the area of the throats [5]. Along the same line of thought, if we pack compressible or deformable grains we need to know the variation of the coordination number to understand the variation of the mechanical properties of the packing. Jernot [3] gave an empirical relation between the compacity of sintered packings of copper spheres and the mean coordination number of these spheres obtained from stereological studies of 2D cuts but his relation does not have a physical basis.

In real packing of grains it is very difficult to consider the grains as undeformable and their position is not defined with enough accuracy to assume point-like contacts. Some historical examples [6, 7] using packings of identical spheres can give a large spread of results depending on the method of measurements. The lower values between 6 and 7 correspond to the number of actual contacts, the upper limit 13.4 established by Dodds [8] represents the maximal number of possible neighbours around one sphere. The value needed for a particular property, such as acoustic or thermal measurements, can be between these two limits or below it in some specific case (In mechanical transport properties Travers *et al.* [9] have shown that only 2 or 3 contacts around one disk really transmit the constraint for a low pressure at 2D instead of 4 the mean coordination number in a disordered 2D packing of disks). For the understanding of this article, we can say that the case of the geometrical arrangement around one sphere is the only one we can estimate by our calculations. We can demonstrate later that we do not need to determine precisely the quality of the contact to estimate the mean coordination number of the packing. If we use soft grains and apply a weak constraint between them, the contact zone or the « overlapping » one is small enough to apply our treatment.

Several theoretical studies based on morphological concepts have been devoted to the problem of relating the number of contacts per unit volume to 2D stereological measurements : Pomeau and Serra [10] have shown that this parameter can be obtained through measurements performed on a random plane section of a large enough packing. Their method relies on making a random 2D section through the packing and studying the distribution of the smallest distances between two neighboring disks which is correlated to the number of contacts per unit volume. The computation is based on the fact that the probability that two spheres are in contact goes to 1 when the distance between the two corresponding disks tends to zero.

Chermant *et al.* [11] checked experimentally the Pomeau and Serra approach on the coordination number of an ordered FCC packing made out of millimeter size plastic spheres. By means of an image processing system, they obtained the distribution of the shortest distances between disks on a random plane section of the FCC packing and found a mean coordination number equal to  $12.0 \pm 0.3$  which coincides with the true value (Z = 12).

Gardner [12] has suggested a new theoretical approach to obtain the coordination number from the information given by the intercepts of a random line drawn through the packing. Her theory is based on the hypothesis that two consecutive intercepts (i.e. parts of the line lying each inside a sphere) separated by a close distance  $\omega$  (called « separator ») belong to two spheres in contact.

The procedure suggested in Gardner's work has two main advantages. First, it makes use of a random line and does not require any determination of shortest distances as in the procedure of Pomeau and Serra [10]. Second, the inversion problem to link 1D and 3D statistics paradoxically turns out to be much simpler than the 2D-3D process which involves a non-algebraic relation between the number of contacts per unit volume and the slope of the cumulative distribution of  $\omega$  at the origin.

In our previous article [13], we have extended and given a first application of the stereological study of E. Gardner [12] to the number of actual contacts between spheres. Using computer-generated ordered and disordered packings of monodisperse spheres we have compared the theoretical results with those obtained by our method. This approach makes use of the distribution of separators of random lines drawn through the packing. The agreement with known results is good even for a relatively small number of lines, this can be useful for experimental studies on unconsolidated monosize sphere packings. This led us to be optimistic about direct extensions of the study to real and sintered packings.

The purpose of this paper is to extend the study of reference [13] to a modelized sintered grain packing. We consider only weak sintering, which is a good starting point for describing

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both non-perfectly punctual contacts and very close neighbors, as explained at the beginning of the introduction. In this limit, calculations are extensions of those of reference [13], very sintered grains need a more detailed knowledge of the pair correlation function and will not be considered here. We use an assembly of monosize slightly overlapping spheres (sintering ratio  $\leq 11 \%$ ) and derive both analytically and numerically the distribution function for true separators (i.e. separators belonging to intersecting spheres), then deduce the number of necks per grain — the analogous of coordination number (Sect. 2). The results are checked on numerical packings (Sect. 3). To minimize the calculation time needed to study monosize 3D packings we have made the studies both on ordered 3D packings and on disordered ones. The difference between these two kinds of packings does not appear in the studies of real contacts but only in the relative position of the nearest neighbours [13]. Complementary results for the packing fraction and average intercept length are given.

#### 2. Theoretical studies.

Let us begin with some generalities on 1D sections : a line cuts a granular medium along an interval called «intercept» and the interval between two consecutive intercepts is a « separator ». Provided the line is long enough and chosen at random, statistical properties for the intercepts may be related to geometrical features of the grain or pore space (e.g. the packing fraction is deduced from the ratio of the total length inside the grain space to the total length of the line). Further information may be obtained when the shape of the grains is known, especially when they are spherical. For example, the coordination number in a monosize packing of hard, touching spheres can be determined from the slope at the origin of the distribution function  $N_{\rm L}(\omega)$  of the separators  $\omega$  [13]:

$$N_{\rm L}(\omega) \approx \frac{\pi}{3} n_{\rm c} \, \omega R \tag{1}$$

$$\frac{z}{2} = \frac{n_{\rm c}}{n_{\rm v}} \tag{2}$$

where  $n_c$ ,  $n_v$  are the number of contacts and the number of spheres per unit volume and z is the coordination number.

This result is the starting point for the study of models for sintered grains, which we consider from now on. This section is divided in 2 parts : we first present the model ( $\S$  2.1), then we derive the distribution function from a biparticle approach ( $\S$  2.2). Finally these studies will be compared to results on numerical packings ( $\S$  3).

2.1 THE MODEL. — The model is that usually described in the literature [2, 5]. We start from an assembly of monosize spheres and contract the intercentre distance by a factor  $(1 - \eta)$ , the radius R being kept constant. Spheres which were initially at a distance r, are now at a distance  $r(1 - \eta)$  apart, and, if  $r(1 - \eta) < 2R$ , they overlap (Fig. 1a). Grains are now truncated spheres, and the limit area between two overlapping spheres is called the *neck*. Several necks may interfere if the sintering ratio  $\eta$  is large enough. In the present paper, we shall avoid this possibility by restricting to  $\eta < \eta_{max} = 1 - \sqrt{3}/2 \approx 0.134$ . The limit configuration for this to happen is represented in Fig. 1b. In this case,  $z(\eta)$ ,  $n_c(\eta)$  and  $n_v(\eta)$  denote the number of necks per grain, the number of necks per unit volume and the number of grains per unit volume respectively. All three parameters increase with  $\eta$ , but relation (2) still holds.

We come now to the 1D cuts. There are some differences with packings of touching spheres, as intercepts may be of several types. In figure 2, the 4 kinds of intercepts in a system

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Fig. 1. — a) Model for sintered spherical grains,  $\eta = \delta / R$  is the normalized ratio of the sintering « process ». b) Limit configuration allowed  $\eta_{max} = 0.134$ . In this case the three spheres have a common overlapping zone.

of two grains (secant or not) are represented. More complicated intercepts mays arise when other grains are involved (Fig. 2e). Besides, two or more necks may intersect (Fig. 1b) but this will not appear here because of the above restriction on  $\eta$ .

The difficulty as to the analysis of the nature of the intercept is overcome by considering separators as they arise only from configurations 2a-2b. The function of interest is the number of separators smaller than  $\omega$  per unit line, say  $N_{\rm L}(\omega)$ . As in (Ref. [13]), it will be convenient to introduce again the distribution function  $N_{\rm L}^*(\omega)$  of the separators for overlapping spheres (the so-called « true » separators). Of course,  $N_{\rm L}^*(\omega) \leq N_{\rm L}(\omega)$ . As we are interested in the small  $\omega$  behavior only, most of the separators we consider arise from overlapping spheres, thus the two functions are equivalent [13]. We shall see that, more precisely, both start linearly with the same slope at small  $\omega$  and

$$N_{\rm L} \approx N_{\rm L}^{*}(\omega)$$
$$\approx \lambda \, \omega R$$

where  $\lambda$  is related to the number of necks and depends on  $\eta$ .

The introduction of  $N_L^*(\omega)$  may seem irrelevant: image analysis deals mostly with  $N_L(\omega)$  as there is no way to decide from a linear cut (or for a planar section) whether we have a true separator or not. On the other hand,  $N_L^*(\omega)$  is easier to handle theoretically, at least for ordered packings as the intercentre distance is exactly  $2R(1 - \eta)$ . It remains, however, to see how it may be useful in the case of a disordered packing. In both cases, the theoretical determination of the number of necks is directly related to the initial coordination number and to  $\eta$  and it is examined in the following subsection.

2.2 THEORETICAL RESULTS. — The complete calculation of  $N_L^*(\omega)$  is similar to that for the identical hard spheres in contact but is not given explicitly here (see the Appendix for the main points of the derivation for small  $\omega$ ). However, some differences appear in the analysis for ordered and disordered assemblies of sintered grains. We consider them here separately. Ordered packings: They are constructed from the four usual ordered arrays of identical

*Ordered packings*: They are constructed from the four usual ordered arrays of identical touching spheres (SC, BCC, HCP and FCC). Provided that  $\eta < \eta_{max}$ , only spheres initially in



Fig. 2. — Types of intercepts that may occur between 2 (a-b-c-d) and 3 grains (e).

contact can overlap when compressed  $(\eta \leq 1)$ ; their intercentre distance is constant and equal to  $2 R(1 - \eta)$ . The number of necks  $z(\eta)$  when  $\eta$  is non zero remains equal to the coordination number z at  $\eta = 0$  (i.e. z = 6, 8, 12, 12), and  $n_c(\eta)$  and  $n_v(\eta)$  are simply related to their value  $n_c$  and  $n_v$  at  $\eta = 0$ :  $n_c(\eta) = n_c/(1 - \eta)^3$ ,  $n_v(\eta) = n_v/(1 - \eta)^3$ . Moreover, small separators always arise from overlapping spheres only and their number is proportional to the number of initial contacts, i.e. to  $n_c$ . Said differently, functions  $N_L^*(\omega)$  and  $N_L(\omega)$  are identical up to the nearest neighbours distance and are proportional to  $n_c$ . thus,

$$N_{L}(\omega) = N_{L}^{*}(\omega) \quad \text{when} \quad \omega < 2 R \left[\sqrt{2(1-\eta)} - 1\right]$$

$$\approx \frac{\pi}{3} n_{c}(\eta) R \omega \left[ \frac{1-4\eta+2\eta^{2}}{1-\eta} + \varepsilon^{3} F(\varepsilon) \right] \quad \text{when} \quad \omega \to 0.$$
(3a)

Here,  $\varepsilon = \sqrt{2 \eta - \eta^2}$  and  $F(\varepsilon)$  are given explicitly in the Appendix. When  $\eta \approx 0$ , we recover the result for spherical grains  $N_{\rm L}(\omega) \approx \frac{\pi}{3} n_{\rm c} R \omega$ . The number of lines which go through the neck and thus must not be taken into account because of the sintering is simply:

$$N^{(0)} = \pi R^2 \eta n_c(\eta) .$$
 (3b)

Disordered packings: In this case, even if  $\eta < \eta_{\text{max}}$  the number of necks per grain in no longer a constant;  $z(\eta)$  increases with  $\eta$  and the initial intercentre distance r for overlapping grains is in the interval from 2 R to 2  $R/(1-\eta)$ ; we still have  $n_v(\eta) = n_v/(1-\eta)^3$  but the

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similar formula for  $n_c(\eta)$  no longer holds. If  $N_L^*(\omega)$  is again the distribution of separators for overlapping grains (true separators),  $N_L(\omega)$  and  $N_L^*(\omega)$  are no longer equal nor is  $N_L^*(\omega)$  proportional to  $n_c$ , even for small  $\omega$  and, in both functions, an extra term must be added which accounts for the pair correlation function near 2 R let us say  $R_V(r)$ , using the notation of reference [14]. It is easy to see that  $N_L(\omega) = N_L^*(\omega) + O(\omega^2)$  as the *r*-integration for the 2 functions differs only on an interval of length  $\omega$ . In the case of slight sintering, an expansion in  $\eta$  is possible and only the knowledge of the pair correlation function near 2 R is necessary. We have

$$n_{\rm c}(\eta) \approx \left[ n_{\rm c} + R_V^{\rm res}(2R) \frac{2R}{1-\eta} \eta + O(\eta^2) \right] / (1-\eta)^3$$
$$N_{\rm L}^*(\omega) \approx \frac{\pi}{3} \frac{R\omega}{(1-\eta)^3} \left[ n_{\rm c} \frac{1-4\eta+2\eta^2}{1-\eta} + R_V^{\rm res}(2R) \frac{2R}{1-\eta} \eta + O(\eta^{3/2}) \right]$$

and the number of lines crossing the neck is :

$$N^{(0)} = \frac{\pi R^2 \eta}{(1-\eta)^3} \left\{ n_{\rm c} + \frac{1}{2} R_V^{\rm res}(2R) \frac{2R}{1-\eta} \eta + O(\eta^2) \right\}$$

where  $R_V^{\text{res}}(2 R)$  is the non singular part of  $R_V(r)$  at r = 2 R; up to the first order in  $\eta$ , relations (3a)-(3b) remain valid.

One must point out immediately that a function such as  $N_L^*(\omega)$  and  $N^{(0)}$  cannot be determined experimentally with high precision. The main problem occurs in the determination of the total length by which numbers of separators will be divided in order to obtain significant numbers per unit length. In image analysis process, we have to eliminate the « mask border » in which the cut of the spheres is not completely defined. In other respects, we have to decide if the total length of a line can be measured with the two spheres at the extremities of it or just half part of them. The answer has been given in reference [13] in which we define a new series of measurements,  $N_L^*(\omega)/N_L$  and  $N^{(0)}/N_L$  where  $N_L$  is the 1D-connexity number i.e. the number of intercepts (or of separators) per unit line and is related to the surface per unit volume

$$S_{v} = n_{v}(\eta) 4 \pi R^{2} \left[ 1 - z(\eta) \frac{\eta}{2} \right]$$
(4a)

by the stereological relation

$$N_{\rm L} = \frac{S_{\rm v}}{4} \,. \tag{4b}$$

Eqs. (3a-3b) are replaced by

$$\frac{N_{\rm L}^{*}(\omega)}{N_{\rm L}} \approx \frac{z(\eta)}{3} \left(\frac{\omega}{R}\right) \frac{1-4\eta+2\eta^{2}}{(1-\eta)(2-z(\eta)\eta)} \left[1+O(\eta^{3/2})\right]$$
(5a)

and

$$\frac{N^{(0)}}{N_{\rm L}} = \frac{z(\eta) \eta}{2 - z(\eta) \eta} \left[ 1 + O(\eta) \right]$$
(5b)

where the geometry of the packing arises only through the average number of necks  $z(\eta)$ . These equations (5a) and (5b) are the simplest way to estimate the mean coordination

number  $z(\eta)$ . By the study of the distribution of the intercepts we can determine also the initial radius R of the grain, and the study of the compacity can give the number of grains per unit volume. So we can use four equations to solve the four initially unknown parameters of a sintered packing : radius R, mean coordination number z, sintering ratio  $\eta$  and the number of grains per unit volume  $n_y$ .

#### 3. Numerical studies.

As explained in 2.1, a straightforward method for getting sintered packings consists first in a construction of a non-sintered packing of monosize spheres, then in a contraction of the distances between the centers by a factor  $(1 - \eta)$ , the radius *R* being kept constant. Ordered sintered assemblies are constructed from the four usual ordered 3D arrays of identical touching spheres (SC, BCC, HCP and FCC).

For the disordered packings, we used numerical 3D packings constructed with a simple algorithm [15]. The spheres are packed, one by one, starting up from the bottom of a given cube. The new sphere is packed with three contacts chosen randomly on the previously deposited spheres. In order to minimize the computation time and the size of the packing required in order to be able to neglect the wall effects, a periodic boundary condition is introduced : all the centers of the spheres must be inside the cube but some of them may overlap so that we decided, for continuity's sake, to make this part reappear on the corresponding opposite border. In our simulation, we could verify that the initial coordination number of the packing was always 6. Each new sphere brings three new contacts and each contact comes from two spheres thus the mean value of the number of contacts around one sphere is Z = 6.

We have realized several different packings (from 1 000 up to 3 000 spheres) within cubes varying from 10 to 15 times the sphere diameter. In these packings we could calculate precisely the density from the radii of the spheres and coordinates of their centers. This initial value fluctuates between 0.590 and 0.605 as compared to the real density of a monosize sphere packing made with glass beads (around 0.62). The « sintering » process for these packings is obtained by a reduction by a factor related to  $\eta$  of all the coordinates of the spheres, while the radius is kept constant.

As everything can be known and calculated directly in numerical packings of spheres, these simulations are used mainly to test the validity of the above formulas in order to apply then later on in actual disordered packings obtained by different compaction procedures.

As said above, we restrict our study to  $\eta < 0.134$  (actually we have used  $\eta \le 0.11$ ). In spite of this restriction, the compacity may be very important; for example in the HCP case, at  $\eta = 0.11$ , the porosity is less than 10 % and we may expect already a good estimate of what may happen in very sintered disordered systems. In all cases, several packings were realized, their size going from  $15 \times 15 \times 15$  to  $60 \times 60 \times 60$ . Random lines were thrown throughout the packings (up to 300 for small packings, 150 for larger ones) to compensate for the small size of the samples and improve the statistics.

The reduced slope at the origin  $N_{\rm L}^*(\omega)/N_{\rm L}$  and the reduced number of « negative » separators  $N^{(0)}/N_{\rm L}$  are plotted in figures 3 and 4 respectively for the ordered packings. They agree fairly well with their theoretical predictions.

These results can permit us to study the disordered packings with the same assumption : the value of  $\omega$  equal to zero is always related to a contact between two spheres and the number of nearest neighbours is small for  $\omega$  near zero.

In figure 5, we have plotted  $N_{\rm L}(\omega)/N_{\rm L}$  and  $N_{\rm L}^*(\omega)/N_{\rm L}$ , for  $\eta = 0.05$ , for a disordered packing; we see that the 2 curves behave in the same way at small  $\omega$  confirming the previous approach. In figure 6, we have plotted  $z(\eta)$  as a function of  $\eta$  which is no longer a constant





Fig. 3. — Plot of the slope at the origin as a function of the sintering ratio  $\eta$  for several types of ordered packings (SC, BCC, FCC, HCP).



Fig. 4. — Plot of the number  $N^{(0)}$  of negative separators as a function of  $\eta$  for several types of ordered packings (SC, BCC,FCC, HCP).

and grows from 6 to approximately 9. This is in agreement with the value 8 for the coordination number found by Bernal and Mason [6] when using painting at the contact points and which includes near contacts within a relative distance of 5%.

As all parameters of such packings are known, it is possible to have estimates of other quantities. The simplest is the average length  $\langle i \rangle$  of one intercept. From the geometrical and stereological expressions for the packing fraction (or compacity) c

$$\dot{c} = n_{\rm v} V$$

and

$$c = N_{\rm L} \langle i \rangle$$



Fig. 5. — Plot of  $N_{\rm L}(\omega)/N_{\rm L}$  and  $N_{\rm L}^*(\omega)/N_{\rm L}$  versus  $u = \omega/2 R$  for  $\eta = 0.05$  and a disordered packing. They have the same slope at the origin, but  $N_{\rm L}^*(\omega)/N_{\rm L}$  saturates more rapidly.



Fig. 6. — Plot of  $z(\eta)$  versus  $\eta$  for a disordered packing. The packing fraction for  $\eta = 0.11$  is approximately 77 %. The value starts around 6 which is the common value for a disordered 3D packings of spheres and increase to 9 which corresponds to the nearest neighbours coordination number.

where  $V = \frac{4}{3} \pi R^3 \left[ 1 - \frac{z(\eta)}{4} \eta^2 (3 - \eta) \right]$  is the average volume of one grain, once the respective part common to two spheres is taken out and  $N_{\rm L}$  is given by (4a-4b), we have simply

$$\langle i \rangle = \frac{4R}{3} \frac{1 - \frac{z(\eta) \eta^2}{4} (3 - \eta)}{1 - \frac{z(\eta) \eta}{2}}$$
(6)

which reduces to  $\langle i \rangle = \frac{4}{3} R$  when  $\eta = 0$ . Again, the knowledge of  $z(\eta)$  only is sufficient and a more detailed geometrical analysis is not necessary. In the same way, knowing  $\eta$  and measuring  $\langle i \rangle$  can give z. Both theoretical (Eq. (6)) and experimental  $\langle i \rangle$  are plotted in figure 7. The agreement is very good. As a by-product, we have compared the two estimates of the packing fraction; here again, the agreement is good.



Fig. 7. — Plot of  $\langle i \rangle$  as a function of  $\eta$  for the same packings.

#### Conclusion.

In this article, we have extended and given an application of a stereological study by Elisabeth Gardner [12] of the number of actual contacts between spheres. Using computer-generated ordered and disordered packings of monodisperse spheres we have compared the theoretical results with those obtained with our method. This approach makes use of the distribution of separators of random lines drawn through the packing. The agreement with known results is good even for a relatively small number of lines, this can be useful for experimental studies either on unconsolidated monosize spheres packings or real and sintered ones.

We can think of several ways to obtain the distribution of « separators ». A first one uses random cuts of packings which have been filled with epoxy resin before cutting in order to keep the spheres in a fixed position. For large enough isotropic materials, random lines thrown in plane should provide the same results as in space. The first numerical experiments made in Caen using this approach seem to confirm this assumption. In some cases one can use directly lines crossing a material. We may think of using an ensemble of transparent spheres into a fluid of the same index and pass a laser beam through it. If the grains or the solvent are fluorescent, one can determine directly the distribution of separators. Another different system would be the use of an ion beam which progressively etches away a composite material [16]. From the temporal distribution of ions emitted, one can reconstruct the concentration profile along one line.

In numerical packings, it is easy to know which separators correspond to a real contact or not. This is not possible any more in experimental sphere packings, even when the grains are identical. A new step then, which corresponds to the practical situation, consists in considering all separators; we then have to take into account the distribution of the distance between any two spheres. We have shown that the slope at the origin can give the same information.

Other extensions may be thought of, namely to assemblies of non-uniform spheres; these may be investigated, initially numerically, provided the information on the length of all intercepts is stored and not only that of the separators. Following the same procedure as in the present work, we are presently studying numerical arrays of two size spheres.

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#### Appendix.

#### Derivation of the formulas (3a)-(3b).

**1.** NOTATIONS. — Let  $\Gamma_1$  and  $\Gamma_2$  be two overlapping spheres with centres  $C_1$  and  $C_2$  at a contracted distance  $C_1 C_2 = r(1 - \eta)(< 2R)$ , O is the middle of  $C_1 C_2$ , D is a random line. Contact O is the origin,  $C_1 C_2$  the X-axis and in the plane perpendicular to  $C_1 C_2$  at O the other two axes are chosen so that the equations for D are

$$Y = \ell$$
  

$$X \sin \theta - Z \cos \theta + d = 0$$
(A1)

where, as in (Ref. [13]),  $\theta$  is the angle between  $C_1 C_2$  and D,  $\ell$  their smallest distance and d the distance of O to the projection of D onto the ZOX plane. The 2 spheres  $\Gamma_i$  (i = 1, 2) have parametric equations in terms of two angles t and  $\alpha$ :

$$X = \pm R(x - \cos t)$$
  

$$Y = R \sin t \cos \alpha$$
  

$$Z = R \sin t \sin \alpha$$
. (A2)

The line D intersects both spheres outside the sintered region provided the 3 following conditions are fulfilled

$$(d \pm Rx\sin\theta)^2 + \ell^2 > R^2 \tag{A3}$$

i)

 $d^{2} + \ell^{2} \cos^{2} \theta > R^{2} \cos^{2} \theta (1 - x^{2})$ 

(existence of intersections)

ii)

(for each sphere, both intersections are on the same side of the plane X = 0).

iii) 
$$d < Rx \cos^2 \theta / \sin \theta$$
 (A5)

(the intersections for the 2 spheres are on different sides of X = 0); we have set

$$x = \frac{r(1-\eta)}{2R}.$$
 (A6)

In the (d, l) plane, conditions (A3)-(A5) determine successively (see Fig. 8)

— the part common to 2 intersecting circles (the «eye »)

— the outside of an ellipse with half axes  $D_0 = R \cos \theta \sqrt{1 - x^2}$ ,  $L_0 = R \sqrt{1 - x^2}$ . When  $\sin \theta < x$  the ellipse is inside the eye; when  $\sin \theta > x$ , it is tangent to the eye in the four points  $\pm \frac{Rx \cos^2 \theta}{\sin \theta}$ ,  $\pm \frac{R \sqrt{\sin^2 \theta - x^2}}{\sin \theta}$ 

— two lines parallel to the  $\ell$ -axis.



Fig. 8. — Zones of the (d, l) plane which are to be taken into account in the estimation of  $N_{\rm L}^*(\omega)$  at small  $\omega$  (black zone): a) when  $\sin \theta < x$ ; b) when  $\sin \theta > x$ .

(A4)

**2.** SINTERED REGION. — The excluded area  $S(r, \theta)$  (= ellipse when sin  $\theta < x$ , ellipse plus the zone inside the tangency points, when  $\sin \theta > x$ ) measures the number of lines  $v_{\rm I}(r)$  per unit line inside the sintered zone when the intercentre distance is r

$$(1 - \eta)^{3} \nu_{\rm L}(r) = R_{\rm V}(r) \int_{0}^{\pi/2} d\theta \, S(r, \theta)$$
  
=  $R_{\rm V}(r) R^{2} \left[ 1 - \frac{r(1 - \eta)}{2R} \right]$  (A7)

where  $R_{\rm V}(r)$  is the pair correlation function of the initial (i.e. non sintered) packing which is defined in the bulk of the text. Notice that, in the case of ordered arrays,  $\nu_{\rm L}(r)$  is proportional to  $\eta$  for  $r < 2 R \sqrt{2}$ .

3. SMALL SEPARATORS. — We come now to small separators. They imply that the abscisse of the intersection points with the spheres are small i.e. the angles t and  $\alpha$  in (Eqs. (A2)) are close respectively to  $t_0 = \sin^{-1} x$  and  $\alpha_0$  (with :  $\cos \theta \tan \alpha_0 = d/\ell$ ), and that  $d^2 + \ell^2 \cos^2 \theta$  is close to  $R^2 \cos^2 \theta (1 - x^2)$ . Up to the first order, the coordinates of the intersections with the 2 spheres are given by (i = 1, 2)

$$X_{i} = \pm \frac{\sin t_{0}}{\mathbf{D}(\pm)} \left[ \Delta \boldsymbol{\ell} \cos \theta \cos \alpha_{0} + \Delta p \sin \alpha_{0} \right]$$
  

$$Y_{i} = \sin t_{0} \cos \alpha_{0} + \frac{\Delta \boldsymbol{\ell}}{\mathbf{D}(\pm)} \left[ \cos \theta \cos t_{0} \mp \sin \alpha_{0} \sin t_{0} \sin \theta \right]$$
  

$$Z_{i} = \sin t_{0} \sin \alpha_{0} + \frac{1}{\mathbf{D}(\pm)} \left[ \Delta p \cos t_{0} \pm \Delta \boldsymbol{\ell} \cos \alpha_{0} \sin t_{0} \sin \theta \right]$$

whence the separator  $\xi = \sqrt{(X_1 - X_2)^2 + (Y_1 - Y_2)^2 + (Z_1 - Z_2)^2}$  is rewritten as

$$R\xi = \frac{x}{\cos \theta \mathbf{D}(-) \mathbf{D}(+)} \left[ \ell^2 \cos^2 \theta + d^2 - R^2 (1-x^2) \cos^2 \theta \right]$$

with  $\mathbf{D}(\pm) = \cos \theta \cos t_0 \mp \sin \theta \sin t_0 \sin \alpha_0$ , the upper sign in all the above formulas corresponding to  $\Gamma_1$ . The condition  $\xi < \omega$  gives the part within an ellipse close to the initial one (see Fig. 8), with half axes

$$D^{2} = D_{0}^{2} + \frac{\omega R}{x \cos \theta} (x^{2} - \sin^{2} \theta)$$
$$L^{2} = L_{0}^{2} + \omega Rx \cos \theta.$$

In the case sin  $\theta < x$ , the new ellipse is larger than the initial one, but is still inside the eye and the contribution is proportional to the area

$$S_1(r, \theta) = \pi (DL - D_0 L_0)$$
  
=  $\frac{\pi \omega R}{2 x} [x^2 - \sin^2 \theta + x^2 \cos^2 \theta].$ 

In the case sin  $\theta > x$ , conditions (A3)-(A5) limit the area in the  $(d, \ell)$  plane to the domain between the two ellipses where (A5) is satisfied (see Fig. 8); we find

$$S_2(r, \theta) = \omega R \left[ \cos \theta \sqrt{1 - x^2 - \cos^2 \theta} + \frac{x^2 + x^2 \cos^2 \theta - \sin^2 \theta}{x} \sin^{-1} \left( \frac{x}{\sqrt{1 - x^2}} \operatorname{cotan} \theta \right) \right]$$
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whence the final result for the number  $N_{L}^{*}(\omega, r)$  of intercepts smaller than  $\omega$  when the initial intercentre distance is r

$$(1 - \eta)^{3} N_{\rm L}^{*}(\omega, r) = R_{\rm V}(r) \left[ \int_{0}^{\sin^{-1}x} d\theta \sin \theta S_{\rm I}(r, \theta) + \int_{\sin^{-1}x}^{\pi/2} d\theta \sin \theta S_{\rm 2}(r, \theta) \right]$$
$$= R_{\rm V}(r) \frac{\omega}{3} \left[ \pi \frac{2 x^{2} - 1}{x} + (1 - x^{2})^{3/2} F(\sqrt{1 - x^{2}}) \right]$$
(A8)

where

$$F(\sqrt{1-y^2}) = \frac{2}{1-y^2} \left[ 1 + \frac{1-2y^2}{\sqrt{1-y^2}} \frac{\cos^{-1}y}{y} \right], \quad F(0) = \frac{8}{3}.$$
 (A9)

The final distribution behaviour at small  $\omega$  and the total number of lines within the sintered region are obtained by a last integration. We have

$$N_{\rm L}^{*}(\omega) = \int_{2R}^{2R/(1-\eta)} \mathrm{d}r \, N_{\rm L}^{*}(\omega, r) \quad \text{and} \quad N^{(0)} = \int_{2R}^{2R/(1-\eta)} \mathrm{d}r \, \nu_{\rm L}(r)$$

where  $R_V(r) = n_c \,\delta(r-2R) + R_V^{\text{res}}(r)$ . For ordered arrays,  $R_V^{\text{res}}(r) = 0$  when  $r < 2R \sqrt{2}$  and we recover the expressions (3a) and (3b). For  $\eta$  small and disordered arrays, they still hold up to the second order.

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