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Geometrical properties of disordered packings of hard disks

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Résumé. — Nous présentons des résultats expérimentaux et théoriques concernant les propriétés d'empilements bidimensionnels de disques. Nous nous sommes particulièrement intéressés à l'étude des mélanges avec distribution de taille des disques. Les propriétés moyennes, telles que compacité ou coordinance, ne dépendent pas de la composition du mélange, contrairement à ce que l'on pourrait attendre au vu des expériences à 3D. Nous montrons l'existence d'un ordre local dans la position relative de grains de tailles différentes ; cet ordre local peut modifier les propriétés physiques de l'empilement. On donne une expression théorique approchée pour la compacité d'empilement 2D compacts. Elle nécessite uniquement la connaissance de l'aire moyenne des quadrilatères du réseau des contacts réels. Pour des empilements désordonnés compacts de disques identiques, on obtient la limite $c = \pi^2/12 \sim 0,822$.

Abstract. — We present experimental and theoretical results for geometrical properties of 2D packings of disks. We were mainly interested in the study of mixtures with disk size distribution which are of more practical interest than equal disks. Average geometrical properties, such as packing fraction or coordination number do not depend on the composition of the mixture, contrary to what would be expected from 3D experiments. We show the existence of a local order in the relative positions of grains with different sizes ; this local order may modify the physical properties of the packing. An approximate theoretical expression for the packing fraction c of 2D close packings is given. It implies the knowledge of the average area of quadrilaterals of the network drawn from the real contacts only. For equal disk disordered packings, it yields the limit $c = \pi^2/12 \sim 0.822$.

Except by numerical simulations, it is difficult to study the geometrical properties of hard sphere packings, particularly if we consider the real contacts between grains, which are important for transport or mechanical properties of granular materials. Therefore, besides its interest for structural modelling in two dimensions, the study of hard disk packings on a plane can be a good tool for easier approach and understanding of the geometry of 3D packings. Such a 2D study is especially interesting when the disk size distribution forbids a regular pavement of the plane : experimental equal disk packings, when they are dense, exhibit ordered domains, then their structure depends on the contruction mode. In this paper we study the effect of disk size distribution on geometrical and structural properties of 2D disordered packings.

1. Order-disorder in 2D hard disk packings.

It is difficult to give a precise and formal definition of a random packing as it would not easily take into account the steric exclusions, which, for compact packings, may lead to long range geometrical correlations [1]. On the other hand, generating points according to a Poissonian distribution law is not difficult. Starting from this remark, Stillinger *et al.* [2] have proposed an algorithm to construct a random packing of 2D equal disks : a set of points are randomly distributed on a plane ; then, considering these points as centres, equal disks are grown. During the expansion process, any couple of overlapping disks is shifted symmetrically along its axis so to realize a tangential contact. Triplets and more complicated overlappings are rearranged following a similar procedure which minimizes the sum of the squares of the displacements. According to these authors, the final result should be a dense random packing and not the dense ordered packing (triangular lattice).

On this basis, in a beautiful experiment, Quickenden and Tan [3] have built equal disk packings in the following way : small disks are put at random on a plane isotropically stretched rubber sheet, in very loose packing. Then, one lets the rubber shrink and a photograph is taken. The sheet is stretched again and the process is repeated n times, the disks at the

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beginning of the $(n+1)$ th cycle are placed at the very place they occupied at the end of the nth cycle. The packing fraction c (i.e. the ratio of the occupied area to the total area) grows linearly with the number of contraction sequences up to the critical value 0.83 ± 0.015. Afterwards, it grows far more slowly, up to 0.907 which is characteristic of the most compact 2D ordered packing. The transition is also marked by a change in the behaviour of the average number z of true contacts per disk (the so called coordinance or coordination number) : at the transition, $z \sim 4$. Quickenden and Tan consider that this transition is like from dense liquid to solid, as the structure remains essentially disordered before this transition. Numerical simulations by Mason [4] using an algorithm analogous to that of Stillinger *et al.* yields results which are qualitatively similar to those by Quickenden and Tan.

Using the radial distribution function at more dilute concentrations, Berryman [5] also obtains by extrapolation the value 0.82 ± 0.02 for this critical packing fraction ; he characterizes it as being the value after which the system necessarily becomes ordered. We also proposed $z = 4$ as the coordination number at this transition [6].

The experiment by Quickenden and Tan shows that the structure of compact disk packings must be studied in terms of order-disorder rather than in terms of randomness. The topological order in solid state physics is most often defined starting from the nearest crystalline structure (the triangular regular lattice here), which is also the most compact. It is then useful to represent the disk packing by a network. Starting from the set of points which are the centres of the disks, one representation is the Voronoi tesselation (Wigner Seitz cells) : one draws the bisecting lines to the lines joining any two centres ; then the smallest convex polygon surrounding a given centre contains all points in the space closest to this centre. The construction is unique and fills the whole space. Another representation is the dual one ; the vertices are the centres of the disks, and they are connected by lines joining nearest neighbours (defined in Voronoi sense). Figure 1 gives an illustration of such equal disk packings. These representations have remarkable topological properties, the most important of which being that the average number of links at one vertex (or average number of sides per polygon in Voronoi tesselation) is constant and equal to 6. In this formalism, Rubinstein and Nelson [1] have studied the phase diagram of a packing of disks with radius R_1 as a function of the number of impurities (disks with radius $R_2 \neq R_1$) and the ratio R_2/R_1 ; they show the existence of a hexatic phase, characterized by an orientation order, between the domain of existence of the crystalline and amorphous phases. Then, a *maximum disorder* packing presents no order beyond the near neighbour cluster of a disk, the word « near neighbours » must be understood in the Voronoi sense.

Another network representation relies on the real contacts : in the totally triangulated lattice above, only lines going through real contacts are retained. This new network consists mainly in polygons, but may exhibit some isolated sites and dead ends : this occurs in the experiment by Quickenden and Tan for low compacities. The network is completely connected only when imposing to the system and to each grain a stability criterion. This criterion may be based on central interactions considerations [7-8] : each disk must be blocked by its nearest neighbours ; this implies at least 3 real contacts for each grain, the angle between two links being less than π. Another criterion considers that, in two dimensional systems, each grain has at least 2 real contacts with its neighbours (2 degrees of freedom) : it is true for the disk packing constructed under gravity, or that following the Bennett algorithm [9] (which is precisely used by Rubinstein and Nelson) according to which each new grain is placed, with at least *two* contacts, at the nearest place to the centre of the system. This last stability criterion prescribes a real coordination number $z = 4$ for the disordered packing realized grain after grain [6] : the packing is isostatic [10]. In the case of a collective construction process, a lower value may be obtained because of local or global arching — collective equilibrium — in the system. We have performed numerical simulations on a regular triangular lattice submitted to « gravity » along a direction which is not one of the three axes of the lattice [11]. Bonds which are not necessary for the equilibrium of the system and of each individual grain are suppressed. These simulations were done on lattices with 50×50 sites ; they give a value 3.45 ± 0.45 for minimal coordinance. This value is not far from the theoretical one (3.416) derived by Uhler and Schilling [7], though the stability criteria are not identical. One may conclude that the contact coordination number for a stable disordered packing lies between 3.40 and 4.

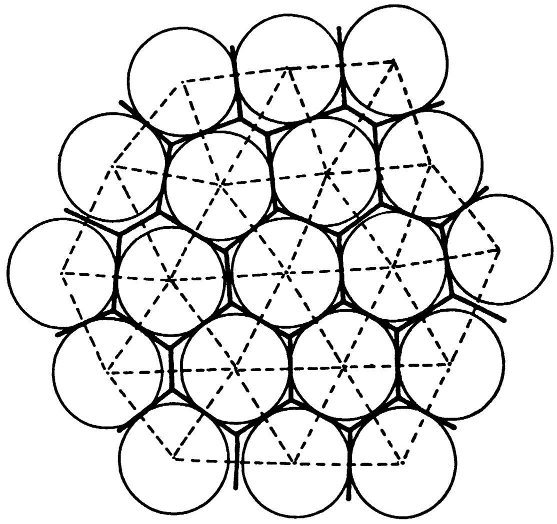

Fig. 1. — Equal disk packing and its triangular network representation. The vertices, or sites, of the network are the centres of the disks. Two neighbour disks are connected by a bond (---). The Voronoi tesselation (full line) is the dual of the triangular network.

A packing with a coordinance equal to or lower than 4 is not necessarily a fully disordered one. As already emphasized in the literature (see for example the review by Berryman [5], packings may include small zones with a regular triangular structure ; in the distribution law for bond angles, this yields an increase of the number of the $\pi/3$ angles. However, an alternative definition of order for equal disks is by reference to the square lattice, because of the peculiar character of coordinance 4 [12]. Figure 2 shows such an example of a packing constructed grain after grain under gravity starting from an irregular wall (a) and its representation by a real network (b), which looks like a distorted square lattice. In such a system, angles between neighbouring links are preferentially equal or near to $\pi/2$. For packings with coordinance close to 4 [12], the average value of the angles is close to $\pi/2$, as quadrilaterals are numerous in the network representation [13]. The angle distribution is then a parameter which characterizes the kind of order of the system. Equidistribution of the angles between $\pi/3$ and $2\pi/3$ should provide a disordered packing, whereas a distribution law favouring angles $\pi/3$ or $2\pi/3$ would indicate a triangular order (6th fold axis), a distribution law favouring angles $\pi/2$ a quadratic one (4th fold axis).

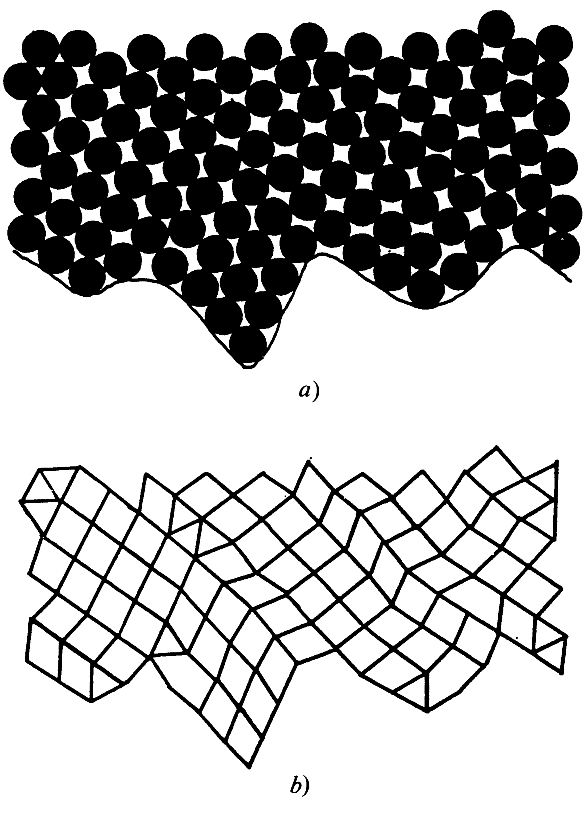

Fig. 2. — a) Equal disk packing built disk after disk under gravity, from a wall of arbitrary shape. b) Representation by a network based on real contacts.

The above constatations have been done mainly by studying equal disk packings. The purpose of this paper is to see how they are modified in mixtures (Sect. 2). The packings with grain size distribution — especially binary packings — may show noticeable differences compared to equal size disk packings. One could think that their mean geometrical parameters, like packing fraction or mean coordination number, vary with the form of the distribution, or with the composition in binary mixtures. We have performed a 2D study. Experimental results are reported in subsection 2.1. They show that, contrary to what would be expected, packing fraction and coordination number are not or not much modified by the grain size distribution. In binary mixtures, this is true only when the ratio of the diameters is not large enough to allow small disks to fit within the cavities of the large grain packing. In subsection 2.2, we give a theoretical expression for packing fraction using only topological constraints. We show that it agrees well with the experimental data (for equal disks, it would lead to the limit $c \sim \pi^2/12 = 0.822$ which is nearly the same as Berryman value [5]).

As for local geometrical properties, like environment of one grain, the analysis is more subtle. Actually, two kinds of disorder may appear. Let us represent the system as a network with real contacts. Either we do not distinguish between small and large disks and the system is topologically disordered ; this was shown by Rubinstein and Nelson [1]. Another possibility is that we distinguish between the two types of centres of disks ; order may appear in the relative positions of the two species, which can be related to the substitution order in the alloys. We shall discuss this further in section 3.

In the conclusion, we discuss how this 2D study could be of use in 3D systems.

2. Packing fraction, coordinance.

This section is devoted to packing fraction and partly to coordination number. The aim is twofold.

a) We first give experimental results for binary mixtures of disks (subsection 2.1). These systems are less studied in the literature though they have a much broader range of practical interest than equal disk packings. Both packing fraction and average coordination number are constant in the whole range of composition and for the studied values of the ratio of large and small disk radii.

b) In subsection 2.2 we propose a theoretical expression for packing fraction. This expression involves some average over polygons of the real contact network. Reasonable statistical assumptions are discussed for equal disk packings. They are supported by numerical experiment. The theoretical formula for packing fraction is also tested for binary mixtures

2.1 EXPERIMENTAL STUDY.

2.1.1 *Experimental conditions.* — Packings with disk size distribution have not much been studied though they intrinsically exhibit topological disorder [1], contrary to equal disk packings. In most cases, their physical properties cannot be described uniquely by means of their average geometrical parameters, like packing fraction or coordination number ; they also depend on the local fluctuations of these parameters, which may be important in case of disk size distribution. Thus, we have performed a systematic study of mixtures with 2 sizes of elements (binary mixtures). The size ratio

$$k = R_1/R_2 \quad (R_1 \geqslant R_2) \tag{1}$$

is taken smaller than 5, in such a way that a small disk cannot enter the cavity formed by three tangent large disks. All our samples are made up of 1 000 disks. The construction recipe is similar to that used by Dodds [14] : a fixed area is filled at random. Then, the system is compressed and vibrated in all directions up to blocking, to prevent anisotropy in the contact directions [15]. These packings are constructed on photographic paper ; we then obtain a photograph « by contact » under indirect lighting, to avoid shadow areas. By doing so, we have a better appreciation of the existence of the contacts because of the contrast between the grain and void phases, as shown in figure 3. Each experimental value is the average value over three packings with the same disk distribution.

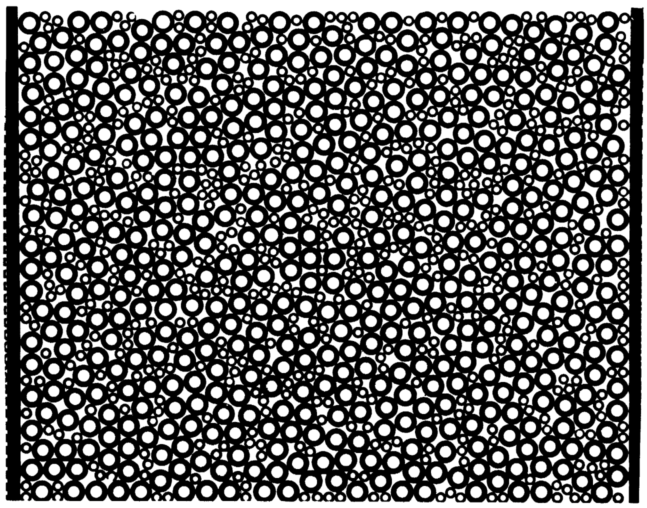

Fig. 3. — Binary mixture of hard disks.

2.1.2 *Experimental results.* — The packing fraction c is a constant within experimental errors [16] for any size ratio $k \leqslant 5$ and any composition

$$c = 0.84 \pm 0.02 . \tag{2}$$

This value is a little higher than the theoretical one (0.82) proposed in next subsection.

The average coordinance $z = \sum n_i z_i$, where z_i denotes the coordinance and n_i the numerical fraction of species i, is also a constant and is equal to

$$z = 3.75 \pm 0.01 \tag{3}$$

for any composition and ratio $k (\leqslant 5)$. This is somewhat smaller than 4, essentially because of the friction between disks and with the construction surface, and not because of the finite size of the samples [17] as value in (3) is obtained solely from disks wich are not at the boundaries of the packings.

We have determined the coordination numbers z_1 and z_2 as a function of n_1 and k for binary mixtures, always under the same experimental conditions. In figure 4, we have plotted z_1 and z_2 *versus* n_1 for $k = 4$. The results agree rather well with those one should get from Dodds statistical geometric model [14] which can be defined by two assumptions :

a) the relative positions of the sites are at random,

b) compared to the totally triangulated lattice, the missing lines (lack of contact between two disks) of the actual packings have been taken away randomly so as to lower the coordination number from 6 to the actual value.

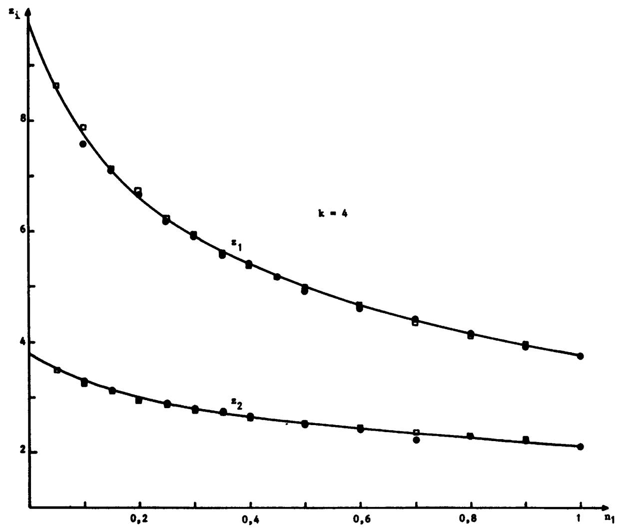

Fig. 4. — Variation of the individual (mean) coordination number of each species in a binary mixture *vs.* the numerical fraction n_1 of large disks. ● : experimental points ; □ : expression (4) ; —— Dodds model.

We have proposed another expression for z_i [11] which takes into account not only geometrical considerations but also the two contact stability criteria. Generally, the average coordination number of one disk in a packing depends on two contributions [18] :

— the equilibrium of the disk under stresses ; for example, a minimum coordinance of 2 is required for stability of disks under gravity (3 for spheres) ;

— the sterical hindrance of the grain.

The coordinance z_i, for maximal disorder (as defined in Sect. 1), may be rewritten as

$$z_i = a + (z - a) \frac{R_i}{R} \tag{4}$$

where $R = \sum n_i R_i$ and $z = \sum n_i z_i$ are the average radius and coordination number ; parameter a corresponds to the (physical) limit of z_i when $R_i \to 0$, which is defined by the equilibrium of the grain only. In the simple case of packings built under gravity, this limit is 2, and is identical to the (geometrical) limit in Dodds model which is not the case for 3 d packing [19] : geometry in 2 dimensions requires a minimal coordinance 3 for a disk with $R_i \ll R$ in a totally triangulated packing. This leads to a real minimal coordinance 2 for a disk in a system with a mean coordinance 4. More generally, parameter a depends on the way the samples are built. In our case, they are constructed on a horizontal plane, and we find $a = 1.75$, value which is comparable to the geometrical limit in Dodds model (1.875) for a packing with a mean coordinance 3.75. So, the relative coordinance z_i remarkably coincides with the experimental value (see Fig. 4).

The same study was carried out with more complicated samples with 10 classes of disks following a truncated lognormal distribution. The ratio k between the radii of the largest and smallest disks was chosen to be less than 5 again. We get the same results as for binary mixtures

i) the packing fraction is equal to 0.84 ± 0.02,

ii) the average coordination number is close to $z = 3.75$,

iii) the dependence of the individual coordinance z_i on R_i is linear, and agrees with equation (4) with $a = 1.75$.

2.2 THEORETICAL EXPRESSION FOR PACKING FRACTION. — In this subsection, we derive a theoretical expression for packing fraction and propose a simple approximation to it (Eqs. (12)-(13)). It is tested on equal disk packings, then on mixtures. We recover the experimental independance on the mixture composition and get for c the value 0.84 ± 0.02. The important point is that the proof requires only the 3 nearly obvious topological relations which describe the real contact lattice, equation (5). This would not work in three dimensions.

2.2.1 *General formula.* — We consider the partitioning of the plane based on real contacts ; stability (at least 2 contacts) implies a fully connected network. The differences with Voronoi tesselation was emphasized in section 1 : z (~ 4) polygons around a site, predominance of triangles and quadrilaterals, fixed length of the edges (sum of the radii of the disks), possibility of non-convex polygons...

The proof is similar to that by Rivier and Lissowski [20] for tissues and froths in the Voronoi representation. Let us denote by N the number of disks, P_n the number of polygons with n sides ($n \geqslant 3$), A_n their average area and E the number of edges. When neglecting the finize size effects, we have the topological constraints

$$\begin{aligned} &\sum_{n \geqslant 3} P_n - E + N = 1 \quad \text{(Euler relation)} \\ &\sum_{n \geqslant 3} nP_n = 2E \qquad \text{(Conservation of the number of edges)} \\ &\sum_{n \geqslant 3} P_n A_n = \text{total given area} . \end{aligned} \tag{5}$$

The average coordinance z is defined by

$$2E = Nz . \tag{6}$$

The packing fraction

$$c = \frac{N \langle \pi R^2 \rangle}{\sum_{n \geqslant 3} P_n A_n} \tag{7}$$

where the brackets $\langle \ \rangle$ stand for the average on all species of disks.

In terms of the probability

$$p_n = P_n \Big/ \sum_{n \geqslant 3} P_n \tag{8}$$

of finding a polygon with n sides and by eliminating E, we can replace the above relations (5) to (7) by

$$\sum_{n \geqslant 3} p_n = 1$$

$$(2 - z) \sum_{n \geqslant 3} np_n + 2z \sum_{n \geqslant 3} p_n = 0\left(\frac{1}{N}\right) \tag{9}$$

$$\sum_{n \geqslant 3} p_n A_n = A_0$$

and

$$c = \frac{2}{z-2} \frac{\langle \pi R^2 \rangle}{A_0} \tag{10}$$

where the r.h.s. of the second equation (9) is negligible when $N \to \infty$ and A_0 is now the average area of a polygon. Eliminating p_3, p_4 in equations (9), we get for A_0 and c respectively the expressions

$$A_0 = A_3 + \frac{6-z}{z-2}\left(A_4 - A_3\right) + \sum_{n \geqslant 5} p_n\left[A_n - (n-3) A_4 + (n-4) A_3\right] \tag{11}$$

$$c = \langle \pi R^2 \rangle \Bigg/ \left\{A_4 + (4-z)\left(\frac{A_4 - 2A_3}{2}\right) + \frac{z-2}{2} \sum_{n \geqslant 5} p_n\left[A_n - (n-3) A_4 + (n-4) A_3\right]\right\} . \tag{12}$$

Up to now, no assumptions have been made about the distributions (p_n) of the polygons, or of their angles ; thus relations (11)-(12) are quite general. In our experimental work, p_n has a maximum value for $n = 3$ or 4 and then decreases, while A_n grows approximately linearly with n as polygons of higher order are rather stretched (see below). The summations in equations (11)-(12) may be neglected as a first approximation. Moreover, as z is not far from 4 ($z \sim 3.75$ in our experimental work [16]) and $A_4 - 2A_3$ is small, a simple approximation to packing fraction is

$$c \sim \frac{\pi \langle R^2 \rangle}{A_4} \tag{13}$$

where only quadrilaterals appear (though p_3 is not zero).

2.2.2 *Packing fraction of equal disk packings.* — We first study the packings of disks with equal radius R. In the real contact representation, the polygons have sides of definite length $2R$. Consequently, there are only $(n-3)$ independent angles in a polygon of order n. The most frequent polygons are (equilateral) triangles, rhombii (with angle α, $\frac{\pi}{3} < \alpha < 2\pi/3$) and pentagons which are necessarily convex.

Statistical assumptions.

We now assume that the angles of the rhombii are equally distributed in the interval $(\pi/3, 2\pi/3)$. This seems reasonable for disordered packings with $z < 4$. As can be seen from the numerical test below, this is nearly true for $z \sim 4$.

For a rhombus, the average area is

$$A_4 = (2R)^2 \langle \sin \alpha \rangle ,$$

with

$$\langle \sin \alpha \rangle = \frac{\int_{\pi/3}^{2\pi/3} \sin \alpha \, d\alpha}{\int_{\pi/3}^{2\pi/3} d\alpha} = \frac{3}{\pi} ,$$

and we get for compacity

$$c \sim \frac{\pi^2}{12} = 0.822 \tag{14}$$

which fits remarkably experimental results [16]. We shall see that the terms neglected in (12) give a positive contribution (mainly because $A_4 - 2A_3 > 0$ as can be easily checked for equal disks), then expression (14) is an *upper bound* for the packing fraction of completely disordered packing for which the upper bound of z is 4 [3, 6]. This limit, obtained with the 2 contact stability criterion is presumably general [5].

We have tried to see how the N topological constraints

$$\sum \alpha = 2\pi \tag{15}$$

around the N sites could modify this result. We have listed all possible configurations with 3, 4, 5 polygons around one given site, one of them at least being a rhombus : there are two configurations with 3 polygons (2 rhombii + 1 pentagon, 2 pentagons + 1 rhombus), 10 with four polygons, 14 with five polygons. We calculated the average area of the rhombus for each configuration and for a uniform distribution law for the angles. We found that $\langle \sin \alpha \rangle$ for a given configuration remains equal to $3/\pi$ in almost all cases with 3 or 4 polygons or is slightly altered [for 4 rhombii, and 3 rhombii + 1 pentagon we got respectively $\frac{3}{\pi} \times 1.0047$ and $\frac{3}{\pi} \times 1.0019$]. The greatest differences appear where there are five polygons around a site, when there is at most one triangle. In that case, the difference does not exceed 5 %. Actually, these configurations are rare, because they imply important ordered domains. We have omitted the possibility of having hexagons, heptagons... but the constraint (15) for the angles restricts them to a definition domain which is the same as for polygons of lower order and the results would not be different.

Numerical justifications.

The realization of a real analog experiment with equal disks is very difficult. So, we have verified our assumptions by generating a close packing of 2 361 equal disks through a Bennett algorithm [9]. As the packing is constructed disk after disk, there is more order than in an actual experiment. However, we have minimized this drawback by performing the growth around an initial seed of larger diameter which favours the disorder [12]. Thus the coordinance is a bit greater than 4 (actually 4.1) and packing fraction $c = 0.83$ is higher than the announced value ; in the network, we get many equilateral triangles (Fig. 5).

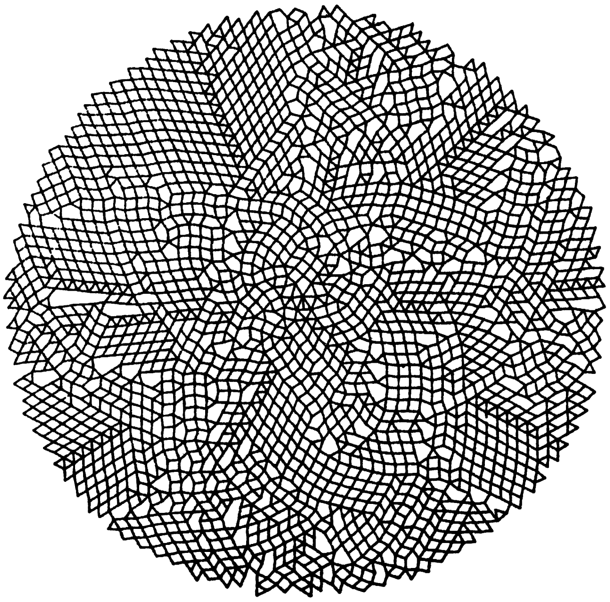

Fig. 5. — Representation by a network of a numerical packing of 2 361 equal disks built according to Bennett algorithm.

The histogram of angles is given in figure 6. We have omitted the 100 first disks and those near the boundary. Except for the predictable peaks at $\alpha = \pi/3$ and $2\pi/3$ arising because of the great number of triangles, the distribution is nearly constant. From the values of p_n given in table I, we can see that the contributions of polygons of order larger than 4 is weak. So the distribution of angles of rhombii is also nearly constant in a large range around $\pi/2$.

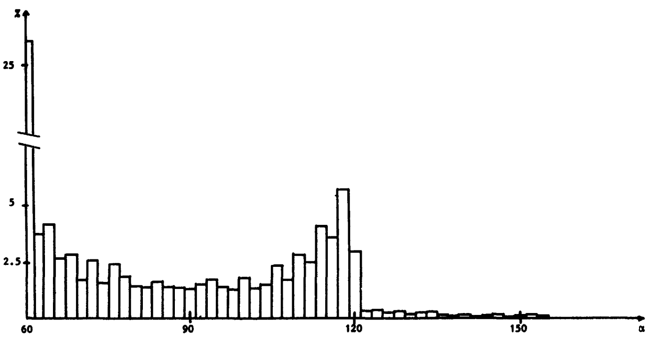

Fig. 6. — Distribution of the angles between two adjacent bonds for the packing of figure 5. In the range 60-120°, the main contribution comes from rhombii. Angles larger than 120° come from higher order polygons.

Finally, we have estimated the frequency of occurrence of the different configurations around one site. For $N = 2\,000$, the results are the following :

i) There are 186 sites with 3 polygons, 1 432 with four polygons, 382 with 5 polygons ;

ii) When there are five polygons around a site, there is at least one triangle ; there are at most 59 sites with one triangle only (i.e. for 3 % of the sites). This is an upper estimate : uncertainty arises because of the impossibility of deciding for 17 sites whether they have one or two triangles. Because of the algorithm itself, configurations with two or more triangles are certainly overestimated as explained above. Nevertheless, the occurrence of situations where $\langle \sin \alpha \rangle$ deviates too much from $3/\pi$ should remain minimal.

It remains to be proven that, under the statistical assumptions discussed above, we may drop in expression (12) for compacity all terms but A_4, so that $c \sim \pi^2/12$.

Table I. — *Analysis of the packing of equal disks obtained through the Bennett algorithm ; n is the number of sides of a polygon, p_n its percentage and A_n its mean area (see Sect. 2.2).*

n	3	4	5	6	7	8
p_n (%)	20.66	70.10	6.11	2.66	0.30	0.17
A_n	0.375	0.835	1.29	1.873	2.434	2.764
$A_n - (n-3) A_4 + (n-4) A_3$	—	—	$< 5 \times 10^{-3}$	0.118	0.219	0.089

First as $A_3 = R^2\sqrt{3}$, $A_4 = \frac{12}{\pi}R^2$, it is easy to see that

$$\frac{A_4 - 2A_3}{2A_4} \sim 0.0466$$

so that the first neglected term is always small and less than 1 % for equal sized disks.

The estimate of the sum

$$\sum_{n \geqslant 5} p_n\left[A_n - (n-3)A_4 + (n-4)A_3\right]$$

is less obvious. From the numerical experiment, we have estimated average areas A_n within an error of 1 % with the help of a digitizer.

Results are reported in table I. Ratio $\frac{A_4 - 2A_3}{2A_4} = 0.0508$ is not very far from the theoretical value. The probabilities p_n decrease very quickly for $n \geqslant 6$ and area A_n increases roughly linearly with n ; then the sum $\sum_{n \geqslant 5}$ yields to A_4 a positive correction of the order of 0.44 % and may be neglected. Thus, approximate expression (13) for c is justified, at least for equal disk packings. As a by-product, let us notice that the p_n do not follow the Rivier-Lissowski law [20]. This proves that other constraints, of sterical nature exist. However, their knowledge is not necessary for the calculation of c because the topological constraints are very strong.

2.2.3 *Compacity of binary mixtures.* — From equation (7) the compacity is

$$c = \frac{2\pi}{z-2}\frac{\sum n_i R_i^2}{A_0}$$

where n_i is the numerical proportion of disks of species i $\left(\sum n_i = 1\right)$, R_i its radius ; A_0 is the average area of a polygon. As for equal disks, we assume $\frac{z-2}{2}A_0 \sim A_4$ i.e.

$$c \sim \pi\frac{\sum n_i R_i^2}{A_4} \qquad (16)$$

and that the average value of A_4 may be calculated by considering that angles are equally distributed within an interval which depends on the length of each side of the quadrilateral.

Starting with a binary packing, we have 6 kinds of quadrilaterals (see Fig. 7). Quadrilateral Q_{1112} may be concave when $R_1/R_2 > \sqrt{2} + 1 = 2.4142$ as a small disk may enter the cavity between four big disks forming a square.

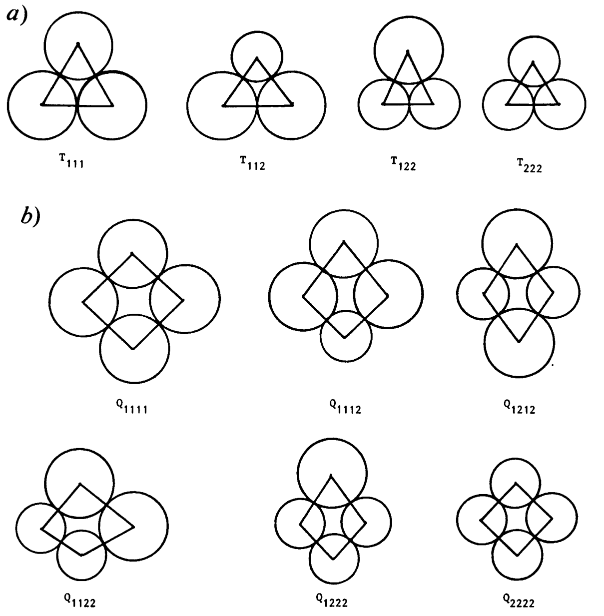

Fig. 7. — Different types of triangles and quadrilaterals in a binary mixture of disks.

We have calculated the average area of each kind of quadrilateral, taking care that the measure which provides entropy invariance [21] is not $d\alpha$, but $(d\alpha + d\beta)$ (where α, β are opposite angles). The theoretical formula is given in table II. We now attribute its correct weight to each configuration. A first idea is to choose the weight related to the numerical proportion n_i of each species, i.e. the binomial coefficients n_1^4, $4n_1^3n_2$, $2n_1^2n_2^2$, $4n_1^2n_2^2$, $4n_1n_2^3$, n_2^4 respectively. The packing fraction is then too large (of the order of 0.94 for $k = 2$ when $n_1 \sim 0.5$). The reason is that the number of contacts 2-2 between two small disks is over-estimated. The ability to have contacts is actually measured by the relative coordinance

$$x_i = n_i z_i / z \qquad (17)$$

which is a function both of the relative sizes and the proportions of the two species in the mixture (Dodds random model). The weights are now

$$x_1^4,\ 4x_1^3x_2,\ 2x_1^2x_2^2,\ 4x_1^2x_2^2,\ 4x_1x_2^3,\ x_2^4\,. \qquad (18)$$

Results are given in table III for $k = 2$ and 4. The packing fraction is remarkably constant and close to its experimental value. Assuming c to be constant and equal to $\pi^2/12$, we get z_i/z as a function of n_i. The curve fits well the experimental results of figure 4. Finally, we calculated the fractions $\left(p_n\right)$ of polygons of order n. As for equal disks, we see that p_n decreases rapidly for $n \geqslant 5$ so that again approximation (13)-(16) is justified. Again, the p_n do not follow the Rivier-Lissowski law, which implies that other sterical constraints exist. The same study was done with mixtures of disks of several sizes, both

Table II. — *General expression for the area A_4 of a quadrilateral generated by four disks with radii R, R', R'', R'''. Centres of the disks with radius R, R''' are opposite sites of the quadrilateral.*

$$A_4 = T_1 + T_2 + 2(X_1 Y_1 - X_2 Y_2)$$

$$T_1 = \frac{1}{2}(R + R')(R + R'')(\cos \alpha_1 - \cos \alpha_2)/(\alpha_2 + \beta_2 - \alpha_1 - \beta_1)$$

$$T_2 = \frac{1}{2}(R''' + R')(R''' + R'')(\cos \beta_1 - \cos \beta_2)/(\alpha_2 + \beta_2 - \alpha_1 - \beta_1)$$

$$X_1 = \sqrt{(R + R')(R + R'')} \cos(\alpha_1/2) \qquad X_2 = \sqrt{(R + R')(R + R'')} \cos(\alpha_2/2)$$

$$Y_1 = \sqrt{(R''' + R')(R''' + R'')} \cos(\beta_1/2) \qquad Y_2 = \sqrt{(R''' + R')(R''' + R'')} \cos(\beta_2/2)$$

with

$$\alpha_1 = \sin^{-1}\left[\frac{2\sqrt{RR'R''(R + R' + R'')}}{(R + R')(R + R''')}\right] \qquad \beta_1 = \sin^{-1}\left[\frac{2\sqrt{R'R''R'''(R' + R'' + R''')}}{(R''' + R')(R''' + R'')}\right]$$

$$\alpha_2 = \sin^{-1}\left[\frac{2\sqrt{RR'R'''(R + R' + R''')}}{(R + R')(R + R''')}\right] + \sin^{-1}\left[\frac{2\sqrt{RR''R'''(R + R'' + R''')}}{(R + R'')(R + R''')}\right]$$

$$\beta_2 = \sin^{-1}\left[\frac{2\sqrt{RR'R'''(R + R' + R''')}}{(R' + R''')(R + R''')}\right] + \sin^{-1}\left[\frac{2\sqrt{RR''R'''(R + R'' + R''')}}{(R'' + R''')(R + R''')}\right]$$

Table III. — *Theoretical packing fraction for different proportions in the binary packing (experimental value* 0.84 ± 0.02). a) $k = 2$; b) $k = 4$.

a)

n_1	0.1	0.2	0.25	0.3	0.35	0.4	0.45	0.6	0.7	0.75	0.8	0.9
c	0.832	0.825	0.832	0.830	0.826	0.830	0.830	0.831	0.823	0.818	0.819	0.824

b)

n_1	0.05	0.1	0.15	0.2	0.25	0.3	0.35	0.4	0.5	0.6	0.7	0.8	0.9
c_1	0.804	0.835	0.832	0.820	0.850	0.830	0.854	0.849	0.834	0.835	0.810	0.837	0.826

when radii have a constant or a lognormal distribution (but truncated in such a way that $k < 5$). Conclusions for packing fraction are unchanged.

3. Order in binary mixtures.

From the agreement with Dodds model for z_1 and z_2, we could deduce that in our samples, the relative position of the disks of each species are randomly distributed. But z_1 and z_2 are mean parameters, i.e. the mean number of contacts around each species. It seems interesting to study experimentally the percentages of the three kinds of bonds t_{12}, t_{11} and t_{22} which, in binary alloy theory are the basic ingredients for the definition of a *local* order parameter (for example Bethe parameter) and may give better information on the structure of the packing than z_1 and z_2.

In particular, the theoretical value for 1-2 bonds in a random packing (in Dodds sense) would be [16] $t_{12}^* = 2\,x_1 x_2$, $x_i = n_i z_i / z$ being the probability that a i-site is the extremity of a bond (def 17). In fact, the experimental values t_{12} are very different. The ratio $\frac{t_{12}}{t_{12}^*}$ varies with the mixture composition as is shown in figure 8, and the maximum value of that ratio is for a composition close to that for which t_{12}^* is maximum. We see that our packings are sterically restricted, even if we can consider that they have no *topological* order when they are described exclusively by an irregular network with bonds between sites. The order we put into evidence by that analysis of hybrid bonds is rather substitution like in this representation.

Going on with the study of one disk environment, we find that the percentages of hybrid triangles and quadrilaterals are greater than would be expected in a random packing. On table V, we have plotted the occurrence frequencies of the different triangles and quadrilaterals (according to the nomenclature of Fig. 7), measured from analog binary samples with $k = 2$ for three different proportions in the mixture. The numbers in parentheses are the percentages in Dodds model. It clearly shows a structural difference

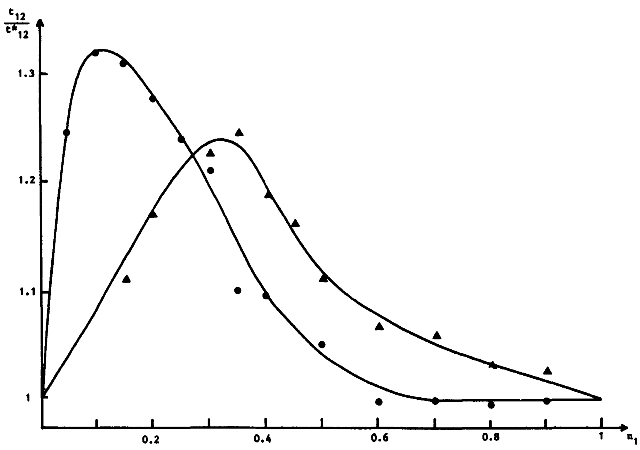

Fig. 8. — Variation of the ratio t_{12}/t^*_{12} of the measured and theoretical (from Dodds model) percentages of 1-2 bonds in binary mixtures, *vs.* the numerical fraction of large disks. ▲ $k = 2$; ● $k = 4$.

Table IV. — *Percentage of polygons with n sides in a binary mixture with $k = 2$, for three compositions of the mixture.*

n_1 \ n	3	4	5	6	⩾7
0.25	44.4	35.5	12.4	4.9	2.7
0.40	38.0	37.7	13.6	7.0	3.7
0.60	39.1	33.9	17.9	6.2	2.9

between analog and random (like Dodds) packings.

The contradiction between that behaviour and the agreement of the z_i's values with random Dodds model is only apparent : this *local* order results from slight collective slips in the lattice, due to the construction mode. We are led to consider that, at least for the ratios k we have studied, the addition of a bond of the $i - j$ type to a disk of species i modifies, by exclusion effect, the environment of this disk : this trends to the suppression of another contact, generally an $i - i$ bond — the coordination number being left constant. Binary mixtures constructed disk after disk under gravity by numerical simulations [22] do not have this local order, as can be seen from inspection of the t_{ij}, up to $k = 4$.

The fact that the mean packing fraction does not seem to be affected by these local displacements is more troublesome. Indeed, one may think that — at least for a size ratio greater than ours, especially greater than the critical $k_c = 6.46$ [6] — this local order leads to a « quasi Appolonian » situation ; small disks go freely into the holes of large disk packings. One should then probably observe a variation of packing fraction as a function of the mixture composition, with a pronounced maximum for a critical composition, like the well-known peak observed in 3D-mixtures [23].

Table V gives also the values for A_4, calculated with the experimental and random weights (last column). The two values are very close to each other, which explains the very good agreement in spite of the structural differences of the two kinds of packings.

Table V. — *Occurrence frequencies — normalized to* 100 — *for triangles and quadrilaterals in a binary packing for several proportions of the mixture.* a) *Triangles*; b) *quadrilaterals. In that case we indicate too the average area A_4 calculated from the experimental and random weights.*

n_1 \ %	T_{111}		T_{112}		T_{122}		T_{222}	
0.10	—	(0.4)	2.90	(5.4)	30.7	(31.6)	66.4	(62.6)
0.25	0.3	(3.6)	22.4	(22.1)	54.80	(44.5)	22.5	(29.8)
0.40	8.28	(12.0)	48.28	(36.9)	36.21	(37.9)	7.24	(13.1)
0.50	18.2	(20.8)	53.8	(42.8)	26.8	(29.6)	0.2	(6.8)
0.60	24.9	(31.8)	56.8	(44.3)	18	(20.7)	0.3	(3.2)
0.75	54.5	(53.2)	41.5	(37.5)	4	(8.8)	—	(0.5)
0.80	59.4	(61.2)	38	(32.5)	2.6	(5.8)	—	(0.5)

b) $R_1 = 1$ $R_2 = 0.5$

n_1 \ %	Q_{1111}	Q_{1112}	Q_{1212}	Q_{1122}	Q_{1222}	Q_{2222}	A_4 exp	A_4 with (18)
0.25 ($x_1 = 0.35$)	— (1.5)	5.1 (11.1)	21.5 (10.3)	18.4 (20.6)	48.2 (38.4)	6.7 (17.8)	1.70	1.71
0.40 ($x_1 = 0.51$)	2 (6.25)	25 (25)	17.8 (12.5)	31.3 (25)	20.6 (25)	1.2 (6.25)	2.06	2.11
0.60 ($x_1 = 0.69$)	14.6 (22.67)	52.4 (40.7)	18.3 (19.15)	10.9 (18.3)	2.8 (8.2)	— (3.7)	2.65	2.72

One open question is to know if that « substitutional » order is uniquely local. For our packings, which are nearly random before compression, and in which compression leads only to local displacement, we think that long range effects, if they exist, will be weak. Our systems are essentially static. It would be interesting to study the same packings under shearing where a longer range order may appear.

4. Conclusion.

The expression we propose for the packing fraction of disordered close packings of hard disks remarkably fits numerical and experimental results, even for packing with grain size distributions. We think that the value $\pi^2/12$ we give for the compacity of *completely* disordered packings of equal disks, is very close to exact value. The corresponding value, for the same order-disorder transition in a 3D packing of equal spheres [5] is close to 0.64. Unfortunately it is not possible to generalize the method of sec 2 to 3D systems since the topological constraints are weaker, and no theoretical expression may be proposed for c in 3D.

Our experimental study indicates a local order in the relative positions of the two species in 2D binary mixtures. This local order does not alter too much the average geometrical parameters, coordinance and packing fraction. But its effects can be important in the physical properties of the system. We shall show in a forthcoming paper that for binary mixtures of disks, it gives rise to variations of the percolation threshold in both cases, when the conducting probability is the same for the two species, and when only one species is conducting.

In 3D packing, for very large ratios of diameters $(k > k_c)$, this kind of local substitutional order becomes segregation when the small grains, if they are in weak concentration, fall at the bottom of the packing, because the void space is connected. But for not too large ratios $(k_c > k > 1)$, we do not have segregation, but only local « order », which may play an important role in the physical or chemical properties, both in the grain and the void spaces. In the grain space, for binary mixtures when the two species have different properties, especially when contacts 1-2 are an essential parameter (solid-solid reaction for example) the effects of that local order can be very important. Even if each species of grains has the same physical properties, local modifications of the connectivity of the system occur both in the grain and void spaces. Perhaps a more important consequence of this local order is to modify the distribution of the distances between grains, which is not studied in this paper. This distribution can be a very critical parameter, as in « Swiss cheese » models for example [24].

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