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Experimental study of radical tessellations of assemblies of discs with size distribution

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Abstract. — We had previously shown that topological and metric properties of 2d mosaics obtained from the Voronoi tessellation of monosize packings of discs deviate from those of a totally random mosaic. Here, we describe a mosaic obtained from the radical tessellation of a two-size mixture of discs at different packing fractions. Two kinds of packings are considered, experimental (discs moving on an air table) and numerical. The deviations are even more striking, particularly at large packing fraction where neither Aboav's nor Lewis' law hold ; moreover, some distributions, such as the distribution of the number of sides of the cells, or distribution of the cell area, are split into two parts, each of them related to one species of discs. Finally, we consider polydisperse packings : as to their topological properties, the mosaics obtained from those packings obey approximately the laws of random mosaics but this is not so for their metric properties, which are still largely affected by steric exclusion.

1. Introduction.

Binary (or more generally, polydisperse) assemblies of discs have been less studied than monosize systems, either from the experimental or theoretical points of view. Most of the results deal with compact assemblies which are supposed to mimic — in the grain space only — bi-dimensional packings under gravity. For the densest (disordered) packing it has been verified experimentally and justified using topological arguments that :

— the maximal packing fraction C_{\max} does not depend much on the percentage of each species and is close to 0.84, a value a little higher than that found in the monosize case ($C \approx 0.82$) [1], and

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— the physical (contact) coordination number z is close to 4 and does not depend much on the mixture either.

Dilute assemblies have been little studied, except for numerical experiments on random sequential adsorption (RSA) assemblies [2]. The development of low dimensionality experiments (adsorption or deposition processes, suspensions of particles, breath figures..) has shown the need for an analysis of disorder at all packing fractions. The notion of « true contact » — which is not always clear even in compact packings — is no longer adequate and must be replaced by the idea of « topological neighbour ». The simplest way for determining the local arrangement around one grain is provided by the radical tessellation proposed by Gellatly and Finney and others [3], which generalizes to polydisperse packings the Voronoï tessellation used for monosize discs or point assemblies [4].

The main aim of this paper is to study binary assemblies of discs with a given size ratio at any concentration and any proportion of small and large discs through the arrangement of the Voronoï polygons. Comparison with « random » mosaics found in the literature and with the mosaics generated from monosize assemblies has been performed and because of the existence of two disc sizes which provides two kinds of cells, many differences are expected.

This paper is essentially *experimental*. The larger part is devoted to the study of binary mixtures of discs with size ratio 2 ; index 1 refers to the small discs and index 2 to the large ones. Most of the discussion is concerned with the results on experimental assemblies of discs realized on an air table already used for studying assemblies of monosize discs [5]. Some numerical packings were built for comparison. The assemblies are described in section 2. The analysis in terms of the radical generalization of Voronoï tessellation is introduced in section 3. Sections 4 and 5 deal with the experimental results for the main indicators of the disorder as arising from the cell analysis : topological properties such as side number distribution, average number of neighbours and first neighbour correlations (section 4), metric properties, typically average areas (section 5). In section 6, we see how these properties are modified when one considers polydisperse packings built numerically.

2. The assemblies.

2.1 EXPERIMENTAL ASSEMBLIES. — The experimental device is described in reference [6]. Because of small local heterogeneities in the porous table, discs move and rearrange permanently. This, together with the absence of friction, allows the realization of homogeneous random assemblies at all densities. Systematic measurements have been performed on binary mixtures ($2 R_1 = 1$ cm and $2 R_2 = 2$ cm) starting from 2 different viewpoints :

- at fixed numerical proportions $n_1, n_2 (n_1 + n_2 = 1)$ of species 1 and 2, we increase the packing fraction C (or concentration, or density) up to the densest disordered packing fraction ;
- at fixed packing fraction C , we change the proportions n_1 and n_2 of small and large discs.

Snapshots of the assembly are taken at different times. They are read using an image processing analysis which determines the position of the centers and the radii. Any new quantity of interest can be derived from these data only. The number of samples is chosen so that we have enough statistics on both species, typically at least a thousand discs in each category, and the errors have been calculated from the dispersion found in measurements on individual photos. Due to constant rearrangements, time averages are also ensemble averages.

2.2 NUMERICAL ASSEMBLIES. — We have tried several kinds of numerical assemblies :

- (1) *At low packing fraction*, we have generated RSA [2] binary assemblies of discs. We recall briefly the procedure : discs are placed one by one, the center being chosen at random. If

the disc does not overlap with already deposited discs, it is definitely placed and does not move anymore ; otherwise, it is rejected and another center is tried, the size being kept. The process ends when a given concentration is reached or when no supplementary disc may be added (jamming limit C_∞). Some variations in the building rule can be used according to the choice of the species of the disc to be placed : one may position all the large discs first and then the small ones — which is easier —, or position the small ones first and then the large ones, which is more difficult, or choose randomly the species of the disc to be positioned. We chose the first alternative.

(2) at the opposite, *for very compact assemblies*, we used a 2d adaptation of the Powell algorithm [7] : grains are placed successively under gravity, in contact with already positioned discs. We get only compact assemblies, with packing fraction of the order of $C_{\max} \approx 0.84$.

For numerical assemblies, some experiments at a larger size ratio ($R_2/R_1 = 4$) and on some polydisperse packings were performed both with RSA and Powell rules. The errors were calculated from the dispersion of the results obtained on several samples ; due to a better statistics (several thousand discs in a numerical packing), they are smaller than in the experimental studies.

3. The procedure.

In compact assemblies, the relative position of discs is correctly described in terms of physical contact. Contacts are no longer interesting quantities in dilute assemblies where only the notion of « neighbourhood » has a meaning. Such an approach was extensively developed in monosize assemblies or point systems : it is the well known Voronoi-Dirichlet tessellation [4], which substitutes the study of the discs by that of their surrounding cells and may be built at any packing fraction, the characteristic cell size scaling with $C^{-1/2}$.

For unequal discs, Voronoi tessellation is not a good choice as it may intersect the large discs. Several generalizations have been proposed and the simplest one, which takes into account the size of each species is the radical tessellation, proposed by Gellatly and Finney [3] . we choose as a separation line between two discs the radical axis, i.e. the line of points with equal tangents (Fig. 1a). This line is outside the two discs and orthogonal to the center line (for touching discs, it is the tangent) ; when the discs are equal, we recover the Voronoi perpendicular bisector. When performing this construction for every pair of discs, polygonal convex cells are generated. Each of them contains one disc and one disc only ; two cells with a common edge are neighbour cells, and the number of sides of a given cell is simply the number of first (or nearest) neighbours for the disc. The hierarchy of first, second, etc. neighbours goes as in the Voronoi case (Fig. 1b). Actually, similarities between mediator and radical lines are deeper : for example, as the vertex coordination number is 3, the topological constraints on 2d graphs yield again an average number of sides $\langle n \rangle = 6$ as for any tessellation.

In the following, the possible correlations between discs are studied through the correlations between the polygons of the radical tessellation. The same analysis was performed both for experimental and numerical assemblies but the results in the paper are quoted mostly for the air table experiments. Results for numerical assemblies (and eventually for a size ratio 4) are given in detail only when they yield supplementary information.

4. Topological properties.

We present here results dealing with the simple counting of the cells and their first neighbours. We follow the same line as in the monosize case and try to see whether the mosaic generated in the radical tessellation is « random » or not. We shall see that many differences arise in the binary case, because of the existence of two different classes of polygons.

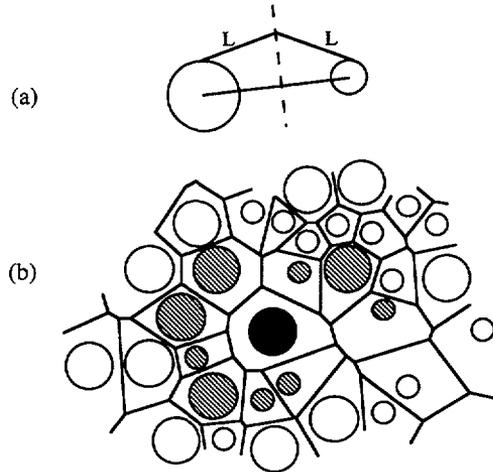


Fig. 1. — (a) Principle of construction of the radical axes. (b) Tessellation in a binary mixture of discs : the striped discs are the neighbours of the black disc.

4.1 DISTRIBUTION OF THE NUMBER OF SIDES OF THE CELLS. — The simplest topological properties can be derived from the knowledge of the distributions $p_n \left(\sum_n p_n = 1 \right)$ and $p_{i,n} \left(\sum_n p_{i,n} = 1, i = 1, 2 \text{ and } p_n = n_1 p_{1,n} + n_2 p_{2,n} \right)$ of the number n of edges of the cells when respectively all discs and discs of species i are considered.

Small and large discs behave differently as density increases. At given proportions and low concentration, the total distribution p_n has a unique maximum near $n = 5$ or 6 ; then at increasing density, it splits into two maxima which get more and more separated. An example for an equal number of discs of each species on the air table is given in figure 2. The distribution is peaked around two values close to $n = 5$ (for small discs) and $n = 7$ (for large discs).

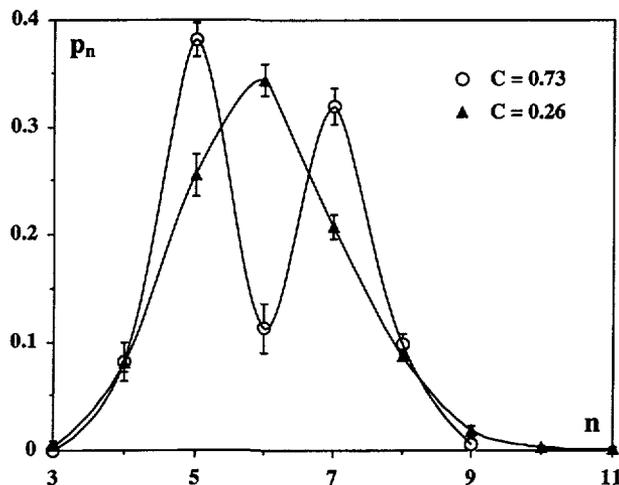


Fig. 2. — Side number distribution p_n for binary assemblies of discs at small and large packing fractions ($n_1 = 0.5$).

The hexagons which were dominant in the monosize case at high compacity are very rare. As a consequence, the percentage p_6 is no longer an order parameter. Although the total variance

$$\mu_2 = \langle n^2 \rangle - \langle n \rangle^2$$

remains high, of the order of 1.5, it can be seen that it does not fit the universal check in terms of p_6 which was put into evidence for the classical random mosaics and in the monosize case [8].

The same holds qualitatively for other concentration ratios on the air table (and for numerical assemblies), i.e. the height and the position of each peak depend on the mixture. For a size ratio 4, and a Powell packing ($n_1 = n_2 = 0.5$, maximum compacity) the separation of the cells into 2 classes is still more striking : 70 % of the polygons around a small disc are quadrilaterals and nearly the same percentage of cells around large discs have $n = 7$ and 8 sides while the total proportion of hexagons is only 7 %.

4.2 ABOAV-WEAIRE LAW. — In classical mosaics, the average number of sides $m(n)$ of the first neighbours of a cell with n edges is well fitted by the empirical law [9]

$$m(n) = 6 - a + \frac{6a + \mu_2}{n} \quad (1)$$

where a is a parameter which is believed to be universal if cells can divide or disappear, and equal to 1 [10]. The Aboav-Weaire law is a first attempt to study the correlations between neighbouring cells : the linear dependence on $1/n$ means that cells with a small number of edges are surrounded by cells with a larger number of sides and conversely. Actually, parameter a is of the order of 0.5 in the Poisson Voronoï tessellations [11] and was found to be an increasing function of the packing fraction in our monosize assemblies [5] (it goes approximately from 0.5 to 1.5). Other counterexamples to the value $a = 1$ were found in breath figures [12] ($a = 2$) and in some evolutive numerical mosaics [13] (where a can be negative !). In these cases, cell disappearance may or may not occur.

In fact, the law (1) appears only as an approximation and some (weak) deviations from the linear dependence of $nm(n)$ on n have been observed numerically [14]. In our binary assemblies, at least at medium or high packing fraction, when the two species are differentiated, the result is more striking : *Aboav-Weaire law does not hold at all*. The variations of $nm(n)$ with n are S-shaped (Fig. 3) ; for $n \leq 5$, most polygons arise from small discs and for $n \geq 7$ from large ones ; a break clearly occurs at $n = 6$ because of the different surroundings of the two classes. Let us note that for $4 \leq n \leq 8$, the values of $m(n)$ come from an average on several hundred cells, and the non-linearity cannot be attributed to a too small statistics.

It is possible to study small and large discs separately. For each species, one gets a linear dependence of $nm_i(n)$ ($i = 1, 2$) with n and $a_1 \approx a_2$ approximately. The a_i 's are increasing functions of the packing fraction and may be large, of the order of 2 or more (when $n_1 = 0.75$ they may reach the value 2.5). However, it is not possible to consider it as a precise result as the linear fit is performed with 3 values of n only for each species.

5. Metric properties.

A study similar to that for monosize packings was performed for *metric* properties and particularly for the cell areas. In the monosize case, some distortions to the empirical laws were observed because of steric exclusion : for instance the Lewis law does not hold.

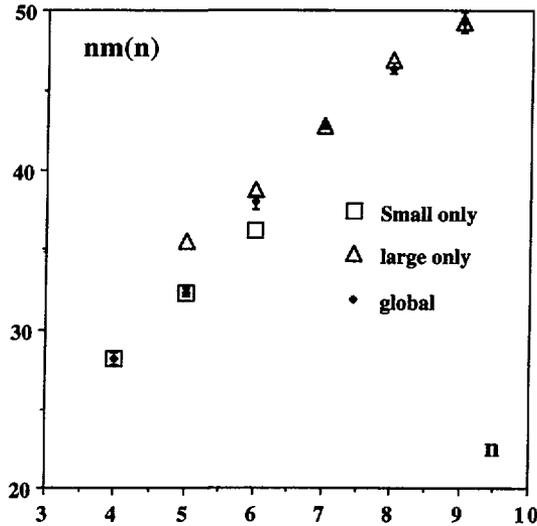


Fig. 3. — Aboav's law is not verified in dense binary packings (here $C = 0.73$ and $n_1 = 0.5$).

Nevertheless, the behaviour of the total area distribution remains rather regular and follows a gamma distribution [5, 15].

In binary mixtures, the differences are more striking : as the radical axis is closer to the center of the small disc, the area of the polygons is directly related to the size of the discs they contain (this is one of the reasons for having chosen such a tessellation). The different behaviour of the 2 classes is more accentuated than in the topological properties.

5.1 DISTRIBUTION OF THE AREAS. — In figure 4, we have plotted the histogram of the normalized areas for an equal number of small and large discs and for the monosize packing at the same density (medium compacity, $C \sim 0.45$) : for the binary assembly, the two populations are already well separated. For higher compacity, the separation is even more marked, the histogram is split into two completely separated parts. Of course, larger size ratios lead to still larger separations.

5.2 LEWIS LAW. — Lewis [16] has shown that in biological tissues, the average area A_n of the cells with n sides is an increasing (linear) function of n .

$$A_n = A_0(n - n_0)$$

where A_0 and n_0 are parameters of the tissue. This law was verified later for most random mosaics in the literature, though a similar law for the perimeter P_n is sometimes a better fit (Desch law [17]). We have shown that the Lewis law and the Desch law do not hold in compact ($C \geq 0.45$) monosize assemblies of discs [5], as A_n (and P_n) exhibits a minimum for $n \approx 5$: steric exclusion is responsible for the high value of the area of low order polygons (triangles including a disc need more place than quadrilaterals and so on..), while the average size of the cells increases again for polygons with 6, 7, etc. sides as all neighbour discs cannot be touching.

In binary mixtures, the strong discrepancy in the disc cell size induces a strong non-linearity of the Lewis law with a rapid change at $n = 6$ (see Fig. 5 for $C = 0.48$, as above ; for $4 \leq n \leq 8$, the values of A_n come from an average over several hundred cells). For a size ratio

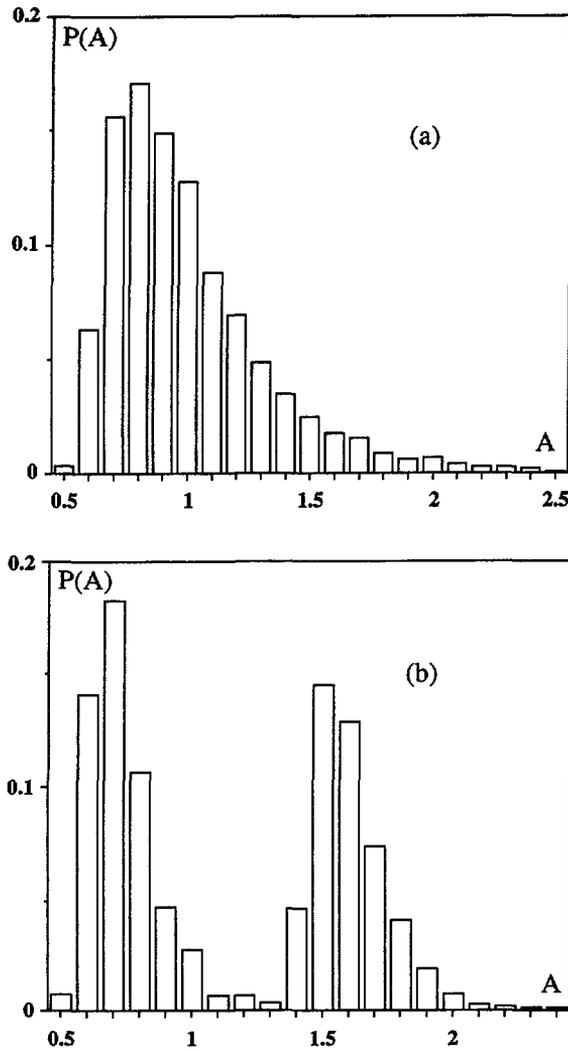


Fig. 4. — Histogram of normalized areas (a) for a monosize packing at $C = 0.46$ (b) for a binary packing at $C = 0.48$ with $n_1 = 0.5$.

of 4 and medium concentration small cells have on average areas of 4 to 10 times smaller than the large ones and the change at $n = 6$ is very sharp. One may wonder whether some linear behaviour may exist separately for small and large discs. Actually this is not so for compact assemblies; if A_n behaves roughly like an increasing (more or less linear) function of n for both sizes at low packing fraction as small and large discs are not very differentiated, in the compact case, the 2 classes behave differently: for small discs, A_n is a function of n similar to that in the monosize case, with a minimum (at $n \approx 4$) due to the steric exclusion. For large discs, A_n is nearly constant or can even slowly decrease with increasing n : this is because in this case a larger value of n corresponds to a larger number of small discs around a large one and a better space filling.

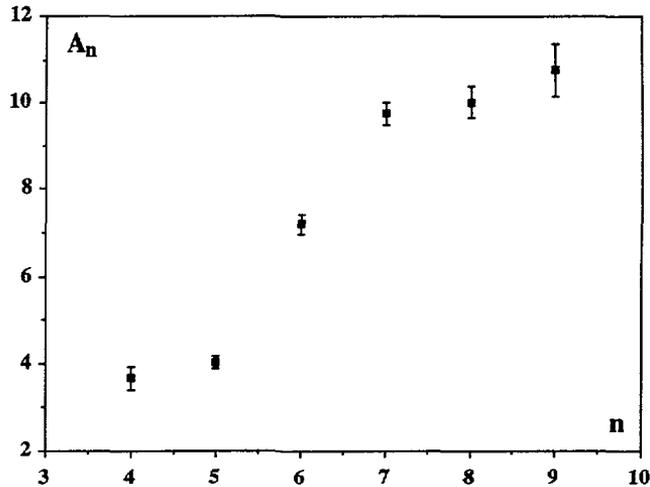


Fig. 5. — Average area A_n as a function of n in a binary packing at $C = 0.48$ with $n_1 = 0.5$: the Lewis law is not verified.

6. Polydisperse packings.

As seen above, at large packing fraction, a binary mixture leads to bimodal distributions and then appears as the more diametrically opposed to the monosize packing. One may then wonder how results are modified when more radius sizes are involved which allow one to get a wide range of cell dimensions. In order to see that, we have built numerical polydisperse packings (6 sizes of discs), either dilute (RSA, $C \sim 0.5$) or dense (Powell, $C \sim 0.85$) packings, with the same values of radii (6, 10, 14, 18, 22, 26) but different proportions of the disc species : uniform, increasing and decreasing linear distributions of the radii.

6.1 TOPOLOGICAL PROPERTIES. — For all these packings, the distribution p_n of the number of sides of the polygons is unimodal with a maximum for $n = 6$ (Fig. 6). Of course, the shape of

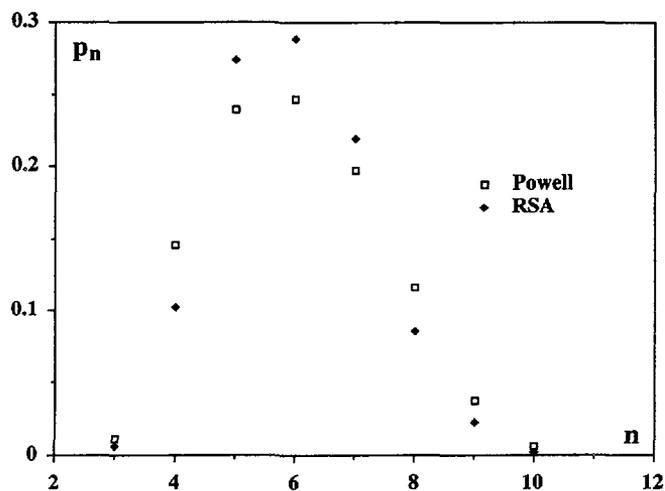


Fig. 6. — Side number distribution p_n in a dense (Powell, $C \sim 0.85$) and a dilute (RSA, $C \sim 0.5$) polydisperse packing for a uniform distribution of radii.

the distribution depends on the composition of the mixture : the more numerous the smallest discs, the wider the distribution for large values of n . At a given packing fraction, the distribution is wider than for a monosize packing and the value of μ_2 is larger. However, the universal law $\mu_2 = f(p_6)$ is well followed by the polydisperse packings.

The Aboav-Weaire law is quite well verified in the polydisperse packings (Fig. 7) with a parameter a close to 1.3. However, the linearity is not as perfect as in monosize packings.

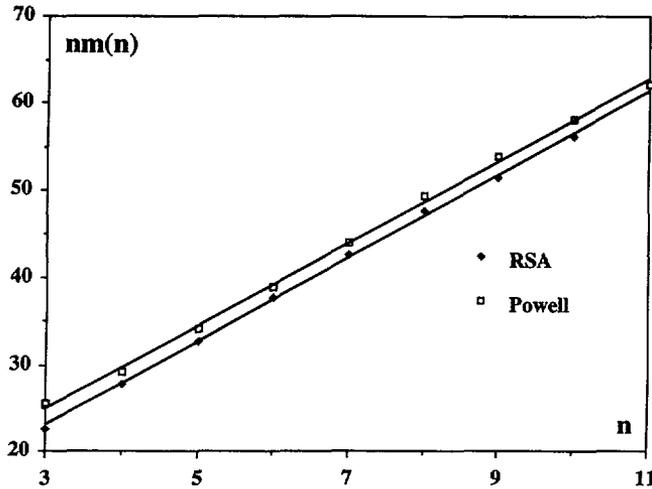


Fig. 7. — Aboav-Weaire law is quite well verified in dense and dilute polydisperse packings (here the radius distribution is linearly decreasing). The errors bars are of the order of magnitude of the symbols.

6.2 METRIC PROPERTIES. — The above results show that the defects appearing in a two-size distribution of discs are to a large part eliminated for a much larger distribution of radii, at least for the topological properties. This is no longer true for the metric properties. Figure 8 shows the histogram of the normalized areas A of the cells for a dense packing with linear increasing

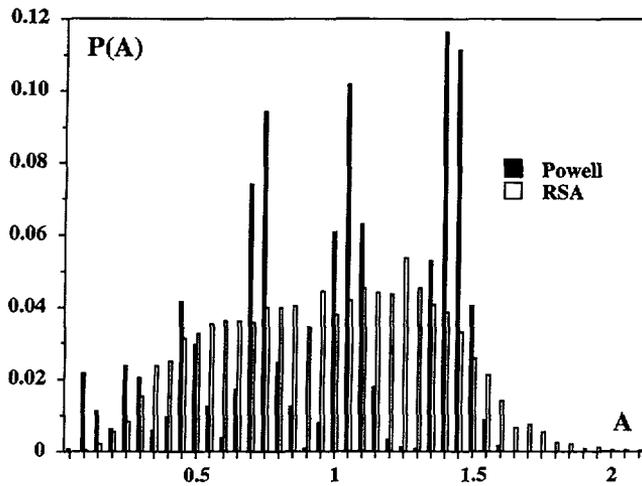


Fig. 8. — Histogram of the area A of the cells, normalized to its average value, for a dense and a dilute packing, in the case of a linearly increasing distribution of radii.

distribution of radii. One can see that the size of the polygons is directly related to the size of the discs : the area distribution is split into 6 separated distributions : each of these comes from one species of discs and its height is reminiscent of the radius distribution. The same is true at large packing fraction for the other compositions. For RSA packings, the histogram of the areas is no longer multimodal as also shown in figure 8, because steric exclusion is less important here.

As can be seen in figure 9, the Lewis law is not verified ; the behaviour of A_n with n is qualitatively the same as in dense binary mixtures (here again, for $4 \leq n \leq 8$, the values of A_n come from an average on several hundred cells, and the non-linearity cannot be attributed to a too weak statistics). The non-linearity is less strong but is still evident in the dilute packings. The values of A_n depend on the disc size distribution : of course, for a given packing fraction, a larger number of small discs leads to cells with smaller size. That explains the relative positions of the curves corresponding to the three radius distributions.

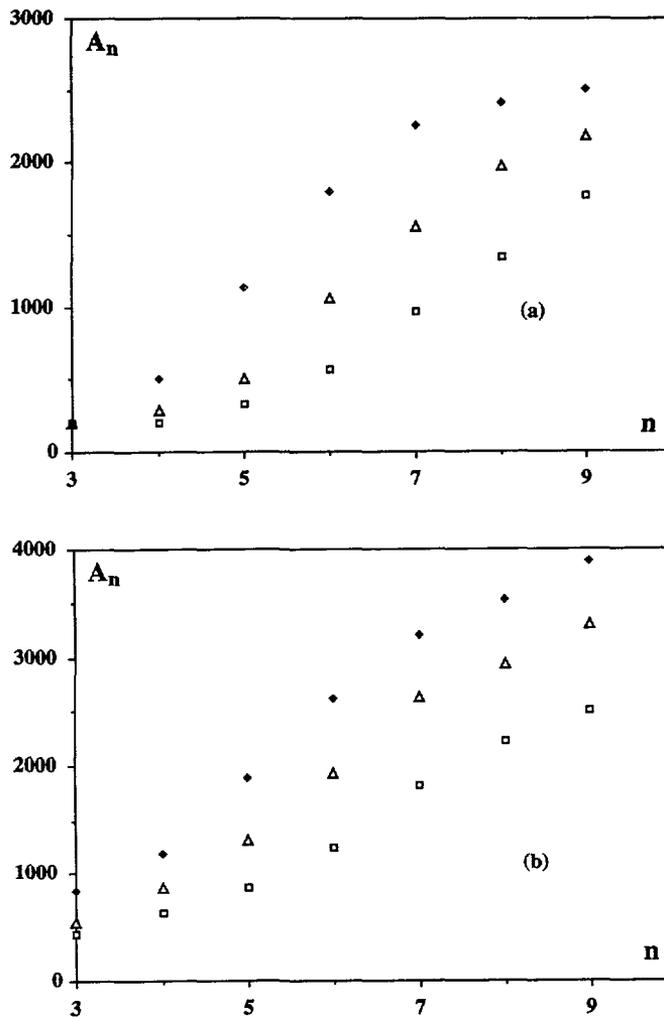


Fig. 9. — Average area A_n as a function of n for dense (a) and dilute (b) polydisperse packings with respectively uniform (Δ), linearly increasing (\blacklozenge) and linearly decreasing (\square) distribution of radii. The Lewis law is not verified.

7. Conclusion.

We have reported here experimental results on the statistics of cells obtained from the radical Voronoï tessellation of binary mixtures of discs.

Mosaics behave very differently from the « random » mosaics in the literature and from the mosaics of the monosize packings. The existence of 2 classes of cells, depending strongly on the size of the central disc (even at medium density) leads to strong non-regularities both in the topological and the metric properties. This holds both for the experimental and numerical assemblies and the effect is more and more accentuated as the packing fraction increases and when the size ratio is large.

Finally, in polydisperse packings, the distribution of the number of sides of the cells is more equilibrated. More generally, as to their topological properties, the mosaics generated from the tessellation of those packings obey approximately the laws of random mosaics. This is not so for their metric properties, which are still largely affected by steric exclusion.

We await with interest the generalisation to binary and polydisperse assemblies of the arguments leading to the linear Aboav [9, 13] and Lewis [10] laws in random mosaics.

Acknowledgments.

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