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A model of binary assemblies of discs and its application to segregation study

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Abstract. — We propose a theoretical model of random binary assemblies of discs which is the extension at any packing fraction of a model previously described by Dodds for dense packings. In dilute assemblies, the notion of contact between the grains must be replaced by that of neighbourhood of the grains. We define the neighbours of a disc through the radical tessellation. The model is tested on packings built grain after grain, obtained from numerical simulations. Its results are then compared with experimental results obtained on assemblies built on an air table: this shows that the possibility of rearrangement in those assemblies leads to segregation at a local scale.

1. Introduction.

It is difficult to prepare mixtures of grains without having segregation. Segregation, which is defined as a non-random spatial distribution of the different species of grains in the medium, has several origins and is influenced by differences in density, shape, interaction and size. Segregation is particularly sensitive to size differences between grains and appears whenever the « history » of the packing allows a relative displacement of one species relatively to the others. Recent experiments on binary mixtures of spheres show that vibrations cause the larger grains to go to the top of the packing (Brazil nuts [1]). In a 2d rotating drum inside which there is a binary mixture of discs, the larger discs go rapidly to the periphery while the smaller ones concentrate at the center of the packing [2]. In 3d, for large size ratios (larger than 6.5-7), small grains may fall under gravity at the bottom of the vessel through the pore space of the large grains, leading to a separation of the two phases.

These are examples of « global » segregation, leading to very inhomogeneous packings. In other circumstances, there may also be a segregation at a more local scale, in which the small

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particles position themselves in a non-random way (for example forming chain-like or compact clusters), although the packing is globally homogeneous. Segregation effects can bring forth serious problems in the use of granular materials because they strongly affect the properties of the materials.

Segregation can be more easily studied in binary mixtures, by looking at the way the particles rearrange according to the dynamics of the packing or the building rule. This has been performed by Bideau [3] for 2d compact static mixtures of discs by examining the local environment of the grains and comparing it with what would be obtained in a random packing such as given by the statistical model of Dodds [4]. No attempt to enlarge the question to dilute assemblies has been performed.

In the present paper, we present a theoretical model for segregation in binary mixtures of discs at any packing fraction. The model is tested both on experimental assemblies of discs moving on an air table [5] and on some packings obtained from numerical simulations. A study similar to that performed by Bideau [3] in compact assemblies has been undertaken, the analysis of contacts being replaced by that of the neighbours. In section 2, we describe the packings. In section 3, we propose a generalization of the statistical model of Dodds for the case of dilute binary mixtures of discs. A local study in terms of percentages of neighbouring cells is presented in section 4.

2. Disc assemblies.

The experimental starting point is an assembly of discs set on a horizontal air table built in Rennes university; most technical and experimental points were already explained in reference [5]. Because of the heterogeneities in the air flux, the discs rearrange permanently and the statistical equilibrium is quickly reached: the assembly is homogeneous at large scale. The experiments were performed with a size ratio 2 (R2/R1 = 2, practically 2 R1 = 1 cm, the index 1 corresponds to small discs and 2 to large ones); the packing fraction goes from C = 10 % to C = 75 % and the ratio n1 of the number of small discs to the total number of discs runs approximately from n1 = 0.2 up to n1 = 0.8. For given packing fraction C and numerical proportions n, of each species (n1 + n2 = 1), several snapshots are taken. Position of the centers and the length of radii are determined through an image treatment analysis and any quantity of interest can be obtained from this information only.

For dilute assemblies, the only way to appreciate without ambiguity whether two discs are neighbours is the radical tessellation proposed by Gellatly and Finney [6] which is the generalization of Voronoi tessellation in monosize packings: each disc is surrounded by a convex polygonal cell which is generated by radical axes and two discs are neighbours if their cells have one edge in common. At one vertex, only 3 lines converge (4 is exceptional and unstable) (Fig. 1). Information on the repartition of the discs is provided by analysing the statistics of the cells, common edges and vertices. The dual of the radical tessellation is a triangular network, generalization of the Delaunay network [7].

Some complementary studies were performed with computer generated packings built grain after grain. Dilute assemblies (C < 50 %) are obtained by a RSA (Random Sequential Adsorption) building rule [8]. Very compact packings are obtained by the Powell algorithm [9] in which each grain is positioned as low as possible, in contact with two already present discs so that the packing fraction is always close to the maximum disordered one, C ≈ 0.84 which is the maximum possible here. In the two algorithms, the species of the disc to be positioned is chosen randomly. We shall see that numerical and experimental assemblies do not behave exactly in the same way.

Finally some RSA and Powell packings were built with an aspect ratio R2/R1 = 4 but no different qualitative conclusions were drawn.

We first define the quantities of interest — with possible generalization to any number of species — then show how they can be derived from a theoretical model which is an extension to any packing fraction of Dodds’ model for dense packings [4] by substituting the neighbours of a given cell to the contacts of the internal disc.

3.1 General quantities. — For a binary packing, and from a topological point of view, three kinds of quantities can be defined at the local level:

a) the (partial) average number $z_i$ of neighbours of a disc of species $i$ ($i = 1, 2$) with the sum rule obtained from Euler’s relation [10]

$$n_1 z_1 + n_2 z_2 = 6$$  \hspace{1cm} (3.1a)

and the (correlated) probability $x_1$ (resp. $x_2$, $x_1 + x_2 = 1$) that a disc of species 1 (resp. 2) is present in a triangle of neighbours. We have

$$z_i = \frac{6 x_i}{n_i}$$  \hspace{1cm} (3.1b)

Of course $x_i \neq n_i$ and as the small species is less present in the triangles, we have $x_1 < n_1$ (resp. $x_2 > n_2$) and $z_1 \ll z_2$.

b) The probability $t_{ij}$ that a disc of species $i$ and a disc of species $j$ are neighbours. We have

$$t_{11} + t_{12} + t_{22} = 1$$  \hspace{1cm} (3.2a)

and

$$x_1 = x = \frac{t_{12}}{2} \quad x_2 = 1 - x = t_{22} + \frac{t_{12}}{2}.$$  \hspace{1cm} (3.2b)

c) The probability $p(ijk)$ for having discs of species $i, j, k$ at the vertices of a triangle. In a binary mixture, there are 4 such quantities and

$$p(111) + p(112) + p(122) + p(222) = 1,$$  \hspace{1cm} (3.3a)
with sum rules

\[ t_{12} = \frac{2}{3} \left[ p(112) + p(122) \right], \]

\[ t_{11} = p(111) + \frac{1}{3} p(112) \]  

\[ t_{22} = p(222) + \frac{1}{3} p(122). \]  

(3.3b)

If we now consider the Delaunay network, the only available quantities are the average distances \( r_{ij}(C) \) between the centers of two discs \( i \) and \( j \) at packing fraction \( C \), or alternatively, the average angles \( \theta_{i}^{(1)}(C) \) with summit \( i \) in a triangle \((i, j, k)\). We are faced to metric quantities depending of course on \( C \) and \((n_1, n_2)\), and in the case of binary mixtures, this means that, on the average, Delaunay triangles are isosceles (triangles \((112)\) or \((122)\)) or equilateral (triangles \((111)\) or \((222)\) with angle \( \pi/3 \)), which was precisely the starting point in Dodds' analysis.

The link between the two networks is achieved by rewriting

\[ z_i = 2 \pi / \theta_i \]  

(3.4)

where \( \theta_i \) is the mean angle around a disc \( i \) in a triangle. We have

\[ \theta_1 = \frac{\pi}{3} p(111) + \left( \frac{\pi}{2} - \alpha \right) \frac{2 p(112)}{3 x} + 2 \beta \frac{p(122)}{3 x} \]  

(3.5)

Angle \( 2 \alpha \) is the mean angle around the site 2 in triangles \((112)\) and \( 2 \beta \) the mean angle around the site 1 in triangles \((122)\). With the above relations, one can write

\[ x = n_1 - \frac{2}{\pi} \left( p(112) \left( \frac{\pi}{6} - \alpha \right) - p(122) \left( \frac{\pi}{6} - \beta \right) \right). \]  

(3.6)

3.2 THE MODEL. — We are now able to set our model at any packing fraction. The argument is divided into 2 steps.

3.2.1 Relation to packing fraction \( C \). — Quantities \( x, t_{ij}, p(ijk) \ldots \) and the angles \( \alpha \) and \( \beta \) depend on the packing fraction \( C \), and it is our aim to find this dependence, at least approximately. Then, we will first try to evaluate the mean distance \( r_{ij} \) between the centers of a disc of type \( i \) and a disc of type \( j \). The mean distance \( r(C) \) between any two neighbour sites depends on \( C \) according to the law

\[ r(C) \sim 1 / \sqrt{C}. \]  

(3.7)

When \( C \sim 0 \), the limits of the quantities of interest are known if we assume the assembly to be random (no interaction between the discs at \( C = 0 \))

\[ \lambda = n_1, \quad z_1 = z_2 = 6, \quad \alpha = \beta = \pi/6. \]

Moreover, the mean distances \( t_{ij} \) between two neighbour sites are all equivalent

\[ t_{11} \sim t_{12} \sim t_{22} \sim t(C). \]

At the other limit of packing fraction, we assume that the packing can be described as a dense ideal packing, in which all the contacts are true (Dodd’s model), i.e. it is made only with equilateral (111 and 222 types) and isosceles (112 and 122 types) triangles. We denote by the
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index \( m \) the quantities related to this packing which has the maximal packing fraction \( C_m \) (this quantity can be easily calculated when the radii of the discs and the composition of the mixture are known). We have:

\[
\begin{align*}
    r_{11m} &= 2R_1, \\
    r_{12m} &= R_1 + R_2, \\
    r_{22m} &= 2R_2.
\end{align*}
\]

The mean distance between any two neighbour sites in this packing is then

\[
    t_m = t_{11m} 2R_1 + t_{12m}(R_1 + R_2) + t_{22m} 2R_2 = r(C_m),
\]

and \( t(C) \) can be written as

\[
    t(C) = r_m \sqrt{\frac{C_m}{C}}. \tag{3.8}
\]

At intermediate packing fractions, let us consider the mean distance \( t_{11} \) between two neighbours of type 1. The ratio \( \frac{r_{11} - t(C)}{r(C)} \to 0 \) when \( C \to 0 \) because its denominator goes to \( \infty \). An approximate form for \( r_{11} \) is the following:

\[
    t_{11} = t_m \sqrt{\frac{C_m}{C} + r_m \frac{R_1 - \langle R \rangle}{\langle R \rangle}} \sqrt{\frac{C}{C_m}}. \tag{3.9a}
\]

In the same way,

\[
    \begin{align*}
        t_{12} &= t_m \sqrt{\frac{C_m}{C} + r_m \frac{R_1 + R_2 - 2\langle R \rangle}{2\langle R \rangle}} \sqrt{\frac{C}{C_m}}, \tag{3.9b} \\
        t_{22} &= t_m \sqrt{\frac{C_m}{C} + r_m \frac{R_2 - \langle R \rangle}{\langle R \rangle}} \sqrt{\frac{C}{C_m}} \tag{3.9c}
    \end{align*}
\]

\( \langle R \rangle \) is the mean radius of neighbour discs at the packing fraction \( C \):

\[
    \langle R \rangle = t_{11} R_1 + t_{12} \frac{R_1 + R_2}{2} + t_{22} R_2. \tag{3.10}
\]

In these expressions, the coefficients of \( \sqrt{C/C_m} \) have been chosen in such a way that

— \( t(C) \) is effectively the mean distance between any two neighbour sites:

\[
    r(C) = t_{11} t_{11} + t_{12} t_{12} + t_{22} t_{22},
\]

— the exact result is obtained for a monosize packing,

— \( t_{11} = 2R_1, \ t_{12} = R_1 + R_2 \) and \( t_{22} = 2R_2 \) for \( C = C_m \).

Of course, the above relations are only approximations that are expected to work all the better since the size ratio is closer to 1. They cannot describe correctly a dense packing with a large size ratio (larger than \( \sim 6.5 \)) and a weak proportion of small discs: in this case, the small discs will be free inside the cavities of the packing of the large ones, and the representation of contacting grains by triangles will be inaccurate. One can note that relations (3.9) imply \( t_{11} + t_{22} = 2t_{12} \), for all the packings we have studied (size ratios 2 and 4, at all packing fractions and compositions) this equality is verified within 1 or 2%.

As already noticed at the beginning of 3.2, the analysis for Delaunay triangles implies that, like for \( C = C_m \), the packing can be modeled as an assembly of equilateral or isosceles
triangles at intermediate packing fractions. Within this approximation, the angles \( \alpha \) and \( \beta \) are given by

\[
\sin \alpha = \frac{r_{11}}{2 r_{12}}, \quad \sin \beta = \frac{r_{22}}{2 r_{12}}.
\]  

(3.11)

where the \( r_{ij} \) are given by equations (3.9). Notice that, in these expressions, parameters \( t_{ij} \) occur implicitly in \( \langle R \rangle \) so that the coupled equations (3.6) and (3.9)-(3.11) cannot be solved without any further information on the conditional probabilities.

3.2.2 Random model. — Equations (3.6) and (3.9)-(3.11) can now be solved if we assume that the packing is random, i.e. if the relative positions of the two species of discs are not correlated. Then all probabilities factorize

\[
t_{11} = x^2, \quad t_{12} = 2 x (1 - x), \quad t_{22} = (1 - x)^2
\]

(3.12)

\[
p(111) = x^3, \quad p(112) = 3 x^2 (1 - x), \quad p(122) = 3 x (1 - x)^2 \quad p(222) = (1 - x)^3.
\]

(3.13)

Equations (3.6) and (3.9)-(3.13) have been solved numerically. They are the basis of our random model. Let us note that all sum rules are obeyed, in particular \( n_1 z_1 + n_2 z_2 = 6 \).

So far, some comments on the relation to Dodds' model are necessary. In Dodds' model, all neighbours are first assumed to be in contact, then some contacts are cut randomly so that the actual coordination number \( z \) satisfies the (approximate) relation \( n_1 z_1 + n_2 z_2 = z \), with experimental \( z \approx 3.5 - 4 \). Actually, here, we keep the Delaunay triangles so that the average number of neighbours is exactly 6 (identity above), which is more satisfactory. The remaining is a generalization at any density, with main assumption in equations (3.9) and (3.11).

4. The small scale segregation.

The random model presented in the previous paragraph is first tested by comparison with the results obtained on the packings obtained from numerical simulations : these packings are built grain after grain and no reorganization is possible, so we expect a small segregation. On the contrary, comparison of the model with the packings built on the air table show a clear segregation in the latter.

4.1 Computer generated packings. — Figure 2 show the variations of \( r_{11}, t_{12} \) and \( t_{22} \) as a function of \( C \) for RSA and Powell packings with a size ratio 2 and equal numbers of small and large discs. The agreement between experimental (on computer generated packings) and theoretical values is fair. For all the packings we have studied, size ratio 2 or 4 and whatever the composition, the discrepancy is less than 5%.

In figure 3, the partial coordination numbers \( z_1 \) and \( z_2 \) are represented as a function of \( n_1 \) for dense Powell packings with size ratio 2. Here again, the agreement with the random model is very good. In figure 4 the same quantities are shown as a function of \( C \) for packings with a size ratio 4 and equal numbers of small and large discs. Again there is a good agreement with the theoretical results. The variations with \( C \) are linear (it is so for all the computer generated packings) and suggest an approximation by a first order development in \( C/C_m \) of equations (3.9) and (3.10), leading to the basic expression

\[
x = n_1 - \frac{\sqrt{3}}{\pi} n_1 (1 - n_1) \frac{R_2 - R_1}{\langle R \rangle_0} \frac{C}{C_m}
\]

(4.1)

with \( \langle R \rangle_0 = n_1 R_1 + n_2 R_2 \), and from which all other quantities can be obtained. This is indeed an excellent approximation.
Fig. 2. — Variation of the $r_{ij}$ with the packing fraction for numerical packings with a size ratio 2 and $n_1 = 0.5$. Full line : theoretical results, (+) experimental points.

Fig. 3. — Plot of the average number of neighbours per species $z_i$ for Powell packings ($C = 0.84$) at increasing proportion $n_1$ of small discs. (O) Theoretical points, (+) experimental points.

Fig. 4. — Plot of the average number of neighbours per species $z_i$ as a function of $C$ for numerical packings with a size ratio 4 and $n_1 = 0.5$. (O) Theoretical points, (+) experimental points.
However the packings obtained from numerical simulations are not totally random because of steric exclusions. Both in RSA and Powell algorithms, it appears to be more difficult to add a large disc than a small one. The small scale segregation can be studied by examining the mean environment of the grains of the two species, in particular the $t_{ij}$. These quantities are compared with the theoretical ones for a size ratio 4 and $n_1 = n_2 = 0.5$ (Fig. 5). As $C$ increases, the small disc cells become smaller and smaller relatively to the large disc cells; $\frac{x_1}{n_1}$ and $t_{11}$ decrease, while $\frac{x_2}{n_2}$ and $t_{22}$ increase. The agreement between experimental and theoretical $t_{ij}$ appears globally correct. However, a segregation is present, as indicated by the experimental values of $t_{12}$, slightly larger than the maximum value 0.5 than can be obtained in a random packing from the relation $t_{12} = 2x(1-x)$. As expected, the discrepancy between theoretical and experimental values is larger for the dense (Powell) packing. In figure 6, we have plotted the $t_{ij}$ as a function of $n_1$ for Powell packings with a size ratio 2. The probabilities

Fig. 5. — Plot of the $t_{ij}$ as a function of $C$ for numerical packings with a size ratio 4 and $n_1 = 0.5$. (O) Theoretical points, (+) experimental points.

Fig. 6. — Variations of the $t_{ij}$ with $n_1$ for Powell packings with a size ratio 2. Full line: theoretical results. (+) experimental points.
$p(ijk)$ of the different types of triangles are also well given by the model (Fig. 7 for a size ratio 2), with the same remarks as to a small segregation. Indeed, the factorizations (3.12) and (3.13) are well obeyed in the packings obtained from numerical simulations. From all these observations, we conclude that our model gives a good representation of a random two-size mixture of discs.

![Graph showing points for different values of $C$ and $n_1$](image)

**Fig. 7.** — Plot of the $p(ijk)$ as a function of $C$ for numerical packings with a size ratio 2 and $n_1 = 0.5$. (○) Theoretical points, (+) experimental points.

### 4.2 Air Table Packings

For the air table packings, comparison with the random model is difficult: at very short distances, discs repel each other so that the air flux can be ejected and there is no true contact between the particles; consequently, in a monosize packing, the regular arrangement of the grains is obtained for a packing fraction less by a factor 1.16 than the maximum value 0.91 for a dense packing of equal discs [11]. To take this effect into account, it is sufficient to divide $C_m$ in all relations of paragraph 3 by 1.16 and multiply $t_m$ by $\sqrt{1.16}$, assuming however that the factor is the same in binary mixtures. All results below are given first at given proportions ($n_1, n_2$) and increasing packing fraction $C$, then at fixed $C$ and increasing $n_1$.

The partial coordination numbers $z_1$ and $z_2$ are plotted as a function of packing fraction $C$ for an equal mixture of small and large discs ($n_1 = 0.5$) in figure 8. The agreement with the model is not as good as in the packings built by numerical simulations. The same study was performed for other proportions of the mixture ($n_1 = 0.75$ and $n_1 = 0.25$). In all cases, the variations are roughly linear, with fluctuations that can be attributed to fluctuations in the composition of the packings for different values of the packing fraction. The large discs have a coordination number larger than in the model. This suggests a larger number of small neighbours for a large disc, in accordance with the structure shown in figure 1, where the chain-like clusters of small discs favours mixed bonds. This is confirmed by the study of the $t_{ij}$. The latter are reported in figure 9 for an equal mixture ($n_1 = 0.5$) at increasing packing fraction. The percentage of mixed neighbours $t_{12}$, much larger than its random limit, 0.5, shows a large segregation. The study of the $p(ijk)$ leads to the same conclusions. As shown in figure 10 for $n_1 = 0.5$, which can be compared with figure 7 for the packing built by numerical simulations, the triangles (112) and (122) are much more numerous than in the random case. The factorizations (3.11) and (3.12) do not hold in these packings.
Fig. 8. — Variations of the $z_i$ with the packing fraction for air table packings with $n_i = 0.5$. (O) Theoretical points, (+) experimental points.

Fig. 9. — Plot of the $t_{ij}$ as a function of $C$ for air table packings with $n_i = 0.5$. (O) Theoretical points, (+) experimental points.

Fig. 10. — Plot of the $p(ijk)$ as a function of $C$ for air table packings with $n_i = 0.5$. (O) Theoretical points, (+) experimental points.
The same is true at constant packing fraction $C$ and increasing $n_1$. As an example, the $t_{ij}$'s are plotted in figure 11 for $C = 63\%$. This figure is to be compared with the same one for Bideau et al. [3] in compact assemblies ($C = 0.84$), built in a collective manner, but where contact percentages were taken into account. Segregation is clear for both air table and Bideau's experiments. On the contrary, Powell assemblies at the same packing fraction $C = 0.84$ yield data closer to that of the random model (Fig. 6). This comforts the idea that segregation may exist in systems where rearrangements are possible and is negligible in numerical models where grains are positioned definitely.

5. Conclusion.

We have presented a theoretical model for the description of the possible geometrical correlations in binary 2d disc assemblies. It is derived from the Dodds model for compact packings, the neighbour replacing the contact, and holds at any packing fraction.

Experimental results show that assemblies built grain after grain by numerical simulations, without reorganization, with size ratio $R_2/R_1 = 2$ behave like random packings at low (RSA) or high (Powell) packing fractions. Even for larger size ratio ($R_2/R_1 = 4$) a first order approximation (Eq. (4.1)) gives good results. On the contrary, assemblies built on the air table show a clear segregation due to the building procedure that allows local reorganization.

In the above analysis, we were interested in very local arrangements of grains, i.e. the mean environment of each type of grains. At an intermediate scale, it seems that small discs rearrange into chain-like clusters along large discs though the packing as a whole remains globally homogeneous. This other aspect of segregation will be studied in a forthcoming paper.

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