ELSEVIER

Powder Technology 93 (1997) 1–11

POWDER TECHNOLOGY

Kinetics of segregation of granular media in a two-dimensional rotating drum

Florence Cantelaube [a,*], Daniel Bideau [a], Stéphane Roux [b]

[a] *Groupe Matière Condensée et Matériaux, URA 804, Université de Rennes I, 35042 Rennes Cedex, France*

[b] *Laboratoire de Physique et Mécanique des Milieux Hétérogènes, URA 857, Ecole Supérieure de Physique et Chimie Industrielles, 10 rue Vauquelin, 75231 Paris Cedex, France*

Received 1 March 1996; revised 5 December 1996; accepted 6 January 1997

Abstract

The segregation of a granular medium in a two-dimensional rotating drum is studied. The particle assembly consists in a bi-modal distribution of disks, with a fixed size ratio of 0.6 and varying relative concentration C. The drum is half-filled with the mixture of grains and it is rotated at a constant velocity Ω so that a permanent surface flow sets in. A very rapid and quasi-complete segregation occurs where the smaller disks gather in a central core. Experimental results on the segregation kinetics are presented for various C and Ω, together with a theoretical analysis of the kinetics of this segregation process based on a description of the surface flow. The segregation can be described as a single-particle process, controlled by a differential convection of the small particles. We show that the steady state is reached exponentially fast in time with a characteristic time constant τ_s, or rotation angle $\Theta_s = \Omega\tau_s$. The segregation time τ_s is predicted to be roughly independent of the concentration in small particles C, and of the rotation velocity Ω. These properties are confirmed experimentally.

Keywords: Granular media; Segregation; Clusters; Kinetics; Time constant; Flow; Differential convection; Percolation; Diffusion; Shearing

1. Introduction

One of the most intriguing and misunderstood phenomena which occurs mostly during granular flows is segregation. When a granular medium is submitted to strain (e.g. by shaking or shearing) the grains having different characteristics (e.g. size, shape, friction coefficient or density) tend to separate spatially from each other. This phenomenon is called 'segregation', and is often observed in industrial processing or in laboratory experiments involving granular media. One of the best known examples is the 'Brazil nut' effect [1–5] in which a very strong particle size segregation takes place. Another example is segregation occurring in inclined chute flow [6,7]. When flowing, the small particles fall to the bottom while the larger ones drift to the top. This is essentially due to a 'percolation' effect and is often observed, for example in natural debris flows [8,9].

Segregation may be a very useful phenomenon, as in mill bowls where the crushing is improved with the particle size separation, but this effect may become troublesome if one wants to obtain a homogenous mixture. A number of recent studies focus on this phenomenon, both for fundamental and applied research. Mixing and segregation are two facets of the same physical process. The underlying mechanisms are diffusion, shearing or convection. The rotating drum gives a good illustration of these two aspects. It is often designed for mixing different particles, but it is a device in which two striking segregation effects are observed: the radial and the axial segregation [10,11]. The radial segregation appears very quickly after the drum starts its rotation, taking the form of a cluster (observable from the lateral sides) of smaller particles along the axis of the drum. Then progressively, a secondary instability sets in which breaks the translational invariance along the axis. Particles separate in more or less regularly spaced bands (alternately formed by large and small particles) if the rotation speed is sufficient: this is axial segregation. Both of these segregation processes are closely connected, and we can wonder if the radial segregation is the initiator of the axial segregation or if, on the contrary, they compete.

In order to study segregation, we have considered a system where only the radial segregation exists by having a two-dimensional geometry. It consists of a two-dimensional circular drum containing rigid and light particles having a simple

* Corresponding author. Present address: UCSD, Department of Applied Mechanics and Engineering Sciences, 9500 Gilman Drive, La Jolla, CA 92093-0411, USA.

PII S0032-5910(97)03213-0

and well-defined shape (disks or pentagons) of macroscopic size. Only frictional contact and inelastic collisions are relevant interactions. This simple model is very well adapted to direct observations and quantitative study from image analysis.

This paper is divided into eight principal sections. The first one is devoted to a description of the experimental set-up; in the second part we describe our kinetics analysis method; then we define in detail a transition operator, and develop a global description of the segregation process after a flow description. In the last part we show how all this theoretical description may be applied to experimental results.

2. Experimental device

2.1. The two-dimensional drum

Our experimental set-up is built from two glass disks of diameter 60 cm, held parallel at their perimeter by an steel annulus. The aperture between these two glass disks is slightly larger than 1 mm. The particles we have used are rigid and light styrene disks 1 mm thick, so the particles can move freely but do not overlap. The experiment is actually in two dimensions. The drum is half-filled with roughly 1400 disks (this number is variable) whose diameter can vary from 6 to 20 mm. The concentration of the smallest particles varies from 0.15 to 0.55 for this study. To avoid the occurrence of geometrical order in our system and slips on the walls, we have irregularly stuck half-disks on the inner outline of the drum. This rough boundary moreover imposed a no-slip condition. The friction between the particles and the wall is very low. Electrostatic effects are nonexistent. The rotation of the drum is driven by a motor and a system of three pulleys, so that the transversal vibrations are minimized. We have changed the rotation speed (Ω) of the drum within a range of 1.3 to 7.2 rpm. The initial homogeneous mixture is achieved by deposing small and large particles randomly in the drum. Then the drum is accelerated from rest and reaches almost instantaneously its rotation velocity for the small values of Ω (less than 4 rpm) considered in this study.

We film the system with a video system and the images are analyzed by an image processing program (Visilog 4.1.4) on a workstation. To obtain well-contrasted images and to simplify the image analysis treatment, all particles are colored in black (large particles) and white (small particles). The images are digitalized in regular time intervals. We have explored different rotation velocities and concentrations of particles. However, we will refer to one particular case which is representative of the entire concentration and velocity range. This 'reference' case is such that Ω is equal to 1.3 rpm, with a system consisting of 600 small (6 mm diameter) and 720 large (10 mm diameter) disks.

2.2. Segregation in the drum

When the drum is filled with disks of different sizes (e.g. with a binary mixture of disks of 6 and 10 mm diameter), segregation occurs: the small disks gather and form a cluster which is located in the middle of the drum. The process is very fast — in less than one revolution, one can observe a nearly complete formation of a segregation cluster.

We show in Fig. 1 two photographs illustrating the initial and steady states of the system: at the beginning of the experiment when the mixture is homogeneous (a) and after one revolution (b). When the drum rotates in continuous-flow regime (i.e. for a rotation speed larger than 1 rpm) the pile is divided into two phases: a 'solid' phase, in the lower part, in which the particles follow the solid rotation of the drum (they do not move in the drum reference frame because of the no-slip condition imposed by the rough boundary) and a 'flowing' phase formed by a few layers of particles flowing at the surface of the solid phase. Since the solid phase is rigidly rotated, the segregation takes place in the flowing phase. We shall consider the details of the convection in the following sections.

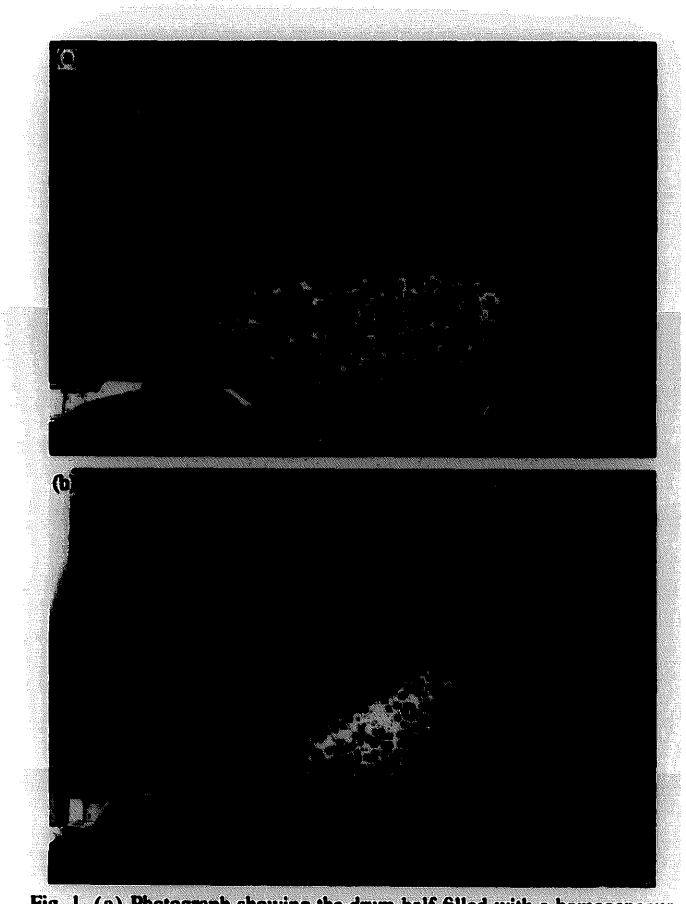

Fig. 1. (a) Photograph showing the drum half-filled with a homogeneous mixture of white (diameter 6 mm) and black (diameter 10 mm) disks before the rotation of the drum. (b) Photograph showing the same mixture after one revolution. A clear segregation is already apparent.

3. Experimental analysis

The first part of our work has been focused on the study of the kinetics of segregation [12]. For clarity we recall the key points obtained in Ref. [12].

3.1. Definition of an order parameter

The homogeneous and the segregated systems are clearly two distinct states of the system. To obtain a quantitative measurement of the degree of segregation, we define an order parameter, P_o, which is by definition equal to unity when the system is segregated and zero when it is homogeneous. Experimentally we use an a posteriori and in situ method to measure P_o. We define a reference cluster, $\mathscr{C}$, which represents an area statistically occupied by small particles in the stationary state, after a large number of rotations [12,13]. The reference cluster is naturally a circular sector in the 'solid' phase. We will see in the section devoted to the surface flow how to 'close' the boundary of the reference cluster, although it concerns only a small fraction of the disks. If the segregation was 'perfect' (either 0 or 100% concentration of small disks) we could define the radius of the reference cluster from the density of the mono-disperse packing. However, since the concentration is only gradually varying we define the reference cluster from the images after a long rotation time. Using image processing software we know the evolution with time of the number of small particles which belong to the reference cluster when the drum is rotating. From this number we calculate the normalized surface, $a(t)$, occupied by small particles, at time t, in the reference cluster. The definition of P_o at time t is then chosen to be

$$P_o(t) \equiv \frac{a(t)-a(0)}{a(\infty)-a(0)} \qquad (1)$$

3.2. The time constant τ

The evolution of the order parameter with time is found experimentally to follow an exponential law:

$$P_o(t) = 1 - \exp\left(\frac{-t}{\tau}\right) \qquad (2)$$

The time constant, τ, characterizes very well the kinetics of the segregation. For example, in the 'reference' case, we find a time constant, τ, equal to (29 ± 6) s. As we have mentioned in the previous subsection, τ is very small: the cluster settles in roughly 0.6 of a drum revolution. From this exponential law we could use the terminology of chemical reaction kinetics, and deduce that this process is of first order. However, we will show that this analogy might be misleading because it suggests that the segregation is a function of the number of disks outside the reference cluster. We will argue that the essential aspects of segregation are captured with a single-particle process.

3.3. Influence of the different parameters on the kinetics

The segregation kinetics has to depend on different experimental parameters: we present in this section the evolution of the time constant first with the concentration of small disks in the mixture, and secondly with the rotation speed of the drum. We show in Fig. 2 the values of the time constant, τ (expressed in seconds), obtained experimentally for different concentrations of small disks: from 0.15 to 0.54. The rotation speed is equal to 1.3 rpm. Each point corresponds to the average value of τ calculated from five different experiments. As can be easily observed in the figure, the time constant does not vary with the concentration, its average value being equal to 32 s. Other experimental results, obtained by Clement et al. [14] who analyze a single tracer trajectory, confirm this observation.

The second experimental result presented here concerns the variation of the time constant, τ (expressed in seconds), versus the rotation speed, Ω, of the drum (expressed in rpm) for a pile composed of 600 small disks of 6 mm diameter and 720 large disks of 10 mm diameter, so that 25% of the pile surface is filled with small disks. The values of Ω vary from 1.3 to 8 rpm. The results are shown in Fig. 3. All the points

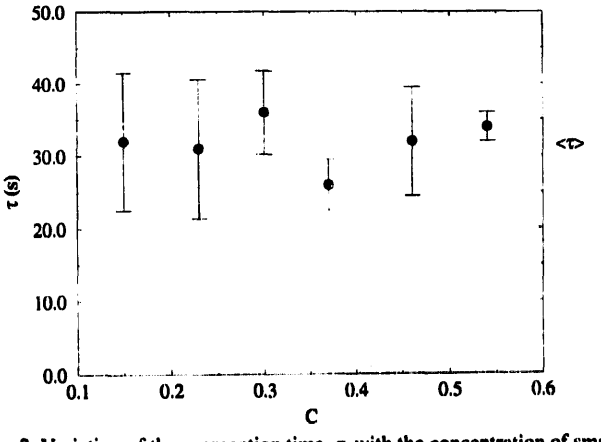

Fig. 2. Variation of the segregation time, τ, with the concentration of small particles.

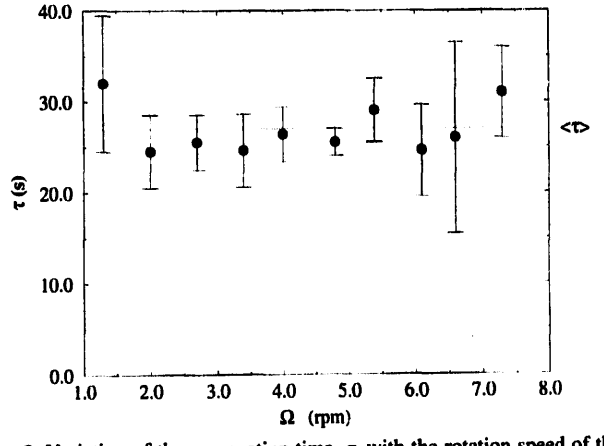

Fig. 3. Variation of the segregation time, τ, with the rotation speed of the drum.

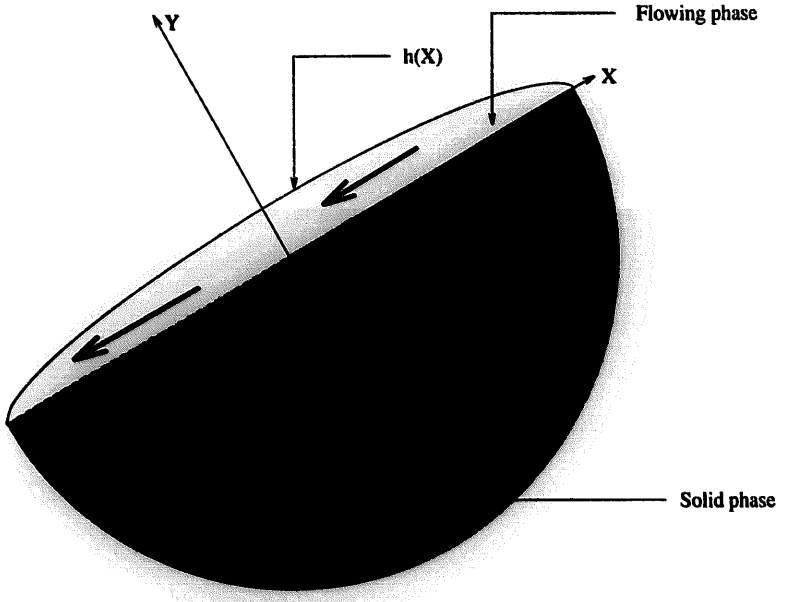

Fig. 4. Two phases in the drum: solid and flowing phases.

are an average calculated from five different experiments. We observe that τ does not depend on the rotation speed.

These two principal experimental results will be explained theoretically in the following sections.

4. Transition operator

Let us focus on a single particle in the assembly. Its motion consists of a succession of two stages. Most of the time, it is part of the 'solid' phase (below the free surface), where the velocity field is strainless. In the experiment, no slip was detected at the wall, so that the motion is a rigid body rotation at the drum velocity. Therefore it simply stays at a fixed distance x from the drum axis. When it reaches the free surface (or is very close to it), it begins to flow with a much higher velocity than in its previous motion. It is finally re-captured by the 'solid phase' and enters the rigid rotation zone, at a presumably different distance from the axis center, as sketched in Fig. 4.

In the case of a half-filled drum, the period T of this cyclic motion is independent of x provided the velocity in the surface flow is much larger than ΩR. It is such that the rotation of the drum is half a revolution, i.e. $T=\pi/\Omega$. In order to study radial segregation, the distance, x, to the axis is the crucial parameter. The simple succession of the two stages in the particle motion suggests considering the discrete ('stroboscope') series of the x value of the particle in the solid phase, x_i, at fixed time intervals of T.

This emphasizes that the dynamics of the mixing is controlled by the recursion relation between x_i and x_{i-1}. This cannot however be expressed as a simple function. Due to the large size of the particles, as compared to the depth of the surface flow, the randomness in the collisions will induce a strongly diffusive motion. Therefore, we may describe the motion of one particle by a *conditional probability* $p(x|x')$, which gives the probability that the particle will be at $x(t+T)=x$ knowing $x(t)=x'$. Let $f_n(x)$ be the probability distribution of x values after n semi-revolutions of the drum that p allows us to compute:

$$f_{n+1}(x)=\int p(x|x')f_n(x')\,dx' \tag{3}$$

The '*transition operator*' $p(x'|x)$ — this term will be justified more precisely — is the key to the description of the motion and, thus, of the segregation in the drum. The rest of this paper is devoted to exploring some properties of p and their relations with the observed experimental results.

The transition probability p reflects the flow pattern of the particles. It thus depends on the nature (here the size) of the particle considered. In our case, two such probabilities have to be studied $p^{(s)}$ and $p^{(l)}$, respectively, for small and large disks. Unfortunately, they depend on the spatial distribution of particles, and thus on time. Indeed as the segregation develops, the details of the velocity field in the flowing region will evolve, and thus so will p. In particular, the diffusion of a small particle in the flow will strongly depend on whether it is surrounded by small or large disks. If the initial distribution of x values for small particles is $f_0(x)$, after n cycles, the distribution becomes $f_n(x)$, which, through iterative application of Eq. (3), can be cast in the following form:

$$f_n(x)=\int_{x0}\int_{x1}\cdots\int_{x_{n-1}} p_n^{(s)}(x|x_{n-1})p_n^{(s)}(x_{n-1}|x_{n-2})$$
$$\ldots p_1^{(s)}(x_1|x_0)\,f_0(x_0)\,dx_0\,dx_1\ldots dx_{n-1} \tag{4}$$

The nature of p as a transition operator appears here explicitly.

Starting from such a general position, we will in the following argue for some simplifying assumptions in order to gain more insight into the physics of the segregation process. First we will argue that the n dependence of the operator can be omitted for the computation of the steady-state properties

and the relaxation time. Then we will constrain the p operator as being the result of a single-particle process, using a 'dilute limit' approximation. Finally to construct the expression of the p operator, we will have to consider first the surface flow field, and the physical mechanism responsible for segregation. This will allow us to compute in particular the relaxation time of the segregation process.

4.1. Long time regime

At late times the system will reach a steady state characterized by a radial distribution of small particles $f_\infty(x)$. Simultaneously, the transition operator will be p_∞. The steady-state property implies that no dependence on the number of cycle will be observed and, thus, using Eq. (3), we obtain:

$$f_\infty(x) = \int p_\infty^{(s)}(x|y) f_\infty(y)\, dy \qquad (5)$$

i.e. the asymptotic distribution f_∞ is a fixed point of the transformation. It can also be seen as an eigenvector of the *linear operator* $\int p_\infty$, with an eigenvalue of unity. The fact that for all y, $p(x|y)$ is a probability distribution imposes that no eigenvalue has a larger modulus. Furthermore, f_∞ is an *attractive* fixed point, as physically expected. The eigenvalues with a real part strictly less than unity will give the rate at which perturbations from the asymptotic regime will decay in time. In particular, the largest of these relaxation times will be of particular interest because it is the dominant mode which can be accessed experimentally. We will come back to this point in detail in the following.

However, from the previous discussion, we have noted that the p operator depends on time, i.e. on the development of the segregation process. This involves a nonlinear character of the transformation since p itself depends on f. This in principle could invalidate the relevance of the previous relaxation times as a basis for estimating the segregation time. If the segregation is an 'auto-catalytic' process, the time for initiating segregation may be the practical limiting process, although once the process is started it might finally relax to its asymptotic value much faster.

From experimental observation, it appears that this is not the case. Initially, the segregation is extremely efficient, and already after one cycle, a very significant fraction of small particles is trapped in the central core of the drum. This observation legitimates the study of the asymptotic operator p_∞ only for extracting the segregation time, i.e. we can linearize the problem in the vicinity of its steady state. Therefore, from the argument developed in this section, we will consider in the following that only the asymptotic operator p_∞ has to be considered, and thus the number of cycle dependence of p can be omitted.

4.2. Dilute limit approximation

We may still simplify the description of the transition operator from the experimental observation of the steady-state distribution of small particles. As can be seen from Fig. 1(b), the concentration of the small particles is high (close to unity) near the central core, and practically zero in the outer zone, with a rather sharp boundary. This observation already leads us to our definition of the order parameter.

Thus, in order to reach this steady state, the key point is to know how a small particle will stay and escape from the outer region, to reach the central core. Indeed, if the small particle is already in the central region, it will have a small probability to escape from it (or otherwise the final state would contain a non-zero concentration of small disks in the outer region). Finally, if the small particle starts in the core and stays in it, its motion will play no part at all in the segregation process.

The important conclusion to be drawn is that the transition operator $p_\infty^{(s)}$ *for any concentration of small disks* can simply be derived from the transition operator computed from the *dilute* case, where only one small disk is present in an assembly of large disks. We thus see that the segregation rate can finally be estimated with an approximation which neglects cooperative effects, which might appear surprising at first glance, although it derives simply from the experimentally observed nearly complete phase separation.

One should not yet conclude from this statement that the segregation time is concentration independent. Indeed, the transition operator at a fixed concentration C can be derived from the one obtained in the limit $C \to 0$, but is not identical. Let x_c denote the radius of the central core where the small particles finally gather. The transition probability at concentration C, $p(x|y,C)$ (we now drop the (s) superscript dealing only with the small disks), has to be written by coarse graining all x smaller than x_c, into a single category which conventionally will be denoted by x_c. We have

$$\begin{aligned}
p(x|y,C) &= p(x|y,0) && \text{if } x < x_c \text{ and } y > x_c \\
p(x_c|y,C) &= \int_0^{x_c} p(x|y,0)\, dx && \text{if } y > x_c \\
p(x|x_c,C) &= \int_0^{x_c} p(x|y,0)\, dy && \text{if } x > x_c \\
p(x_c|x_c,C) &= \int_0^{x_c}\int_0^{x_c} p(x|y,0)\, dx\, dy && (6)
\end{aligned}$$

In general, this coarse-graining does affect the spectrum of eigenvalues of the operator, and thus the relaxation time constants. However, if the eigenvector $f_{slow}(x)$ giving the longest relaxation time of the dilute case is mostly located in the outer region, i.e.: $\int_0^{x_c} f_{slow}(x)\, dx \ll 1$, we can state that the relaxation time will be concentration independent. This can also be used in order to estimate the range of concentrations in which the relaxation time is comparable to the one obtained in the dilute regime. In practice, this range can be fairly large if the slowest process is the escape from the external boundary $x \approx R$ of the drum. We will see that this case is the most relevant one.

5. Flow field

In order to proceed one should have more insight into the flow field. We thus first propose a simple description of the flow. Moreover, as argued above, in the dilute concentration regime, the motion of the small particles will not affect the mean velocity field, and thus we will initially assume that the assembly is mono-disperse.

Let us assume that the flow is friction dominated, and inertia is unimportant. In our experimental system, the disks were light and with a rather low coefficient of restitution, so that, although the flow is evidently gravity driven, the disks reach an equilibrium velocity in a very small distance.

We propose to model the flow by introducing a fixed shear rate γ, uniformly over the entire flowing region. Due to the small thickness of the flowing region [13] (from 3 to 6 diameters of large particles within a Ω range of 1.3 to 5 rpm), and the fact that the free surface is almost flat, this approximation appears reasonable. This shear rate is expected to be only weakly dependent on the slope of the free surface, and thus on the rotation speed of the drum, Ω. The reason for this property is to be found in the observation that, for rotation velocities which are too small, the flow ceases to be continuous, and rather proceeds by avalanches. This tends to indicate that the flow rate cannot be arbitrarily small but rather jumps discontinuously from zero to a finite value. The *relative* evolution of this shear rate with the rotation speed, $(1/\gamma)(d\gamma/d\Omega)$, past the onset of a continuous flow is thus expected to be small. This argument should only be used for a small interval of variation of rotation speeds. However, experimental observations show that a constant γ is a fair approximation in the entire range of considered velocities.

With this assumption, the thickness, $h(x)$, of the flow zone as shown schematically in Fig. 4, results from a conservation law. Let us call y a coordinate perpendicular to x, whose origin is at the interface between the solid region and the flowing zone. For any point in the flowing region, the velocity is parallel to the x axis and its modulus is γy. The flow $\Phi(x)$ in the flowing region at abscissa x is balanced by the incoming flux of particles from the solid region due to the rotation of the drum. Let us also assume that the solid region has a bulk density c_s while the flowing region is dilated to accommodate the flow rate γ, and hence assumes a constant value of c_f in the flowing region. Quantitatively we have

$$\Phi = c_f\gamma \int_0^{h(x)} y\,dy = \left(\frac{c_f\gamma}{2}\right)h(x)^2 = c_s \int_x^R \Omega x'\,dx'$$

$$= \left(\frac{c_s\Omega}{2}\right)(R^2 - x^2) \qquad (7)$$

Hence:

$$h(x) = \left(\frac{c_s\Omega}{c_f\gamma}\right)^{1/2}(R^2 - x^2)^{1/2} \qquad (8)$$

The prefactor of the square root in the above expression is denoted as η in the following. In practice the distinction between c_s and c_f is expected to be a few percent and, thus, as a first approximation, it can be neglected so that $\eta^2 \approx (\Omega/\gamma)$.

6. Segregation mechanism

With this description of the mean granular flow, we may now address the description of the segregation process itself, i.e. the relative motion of a small particle immersed in the mean flow. The velocity of the small particle is equal to $U+v$ where U is the velocity of the large particles. We assume that v is small compared to U. The velocity v has longitudinal and transverse components. We can neglect the longitudinal one (along x) since $v \ll U$, and thus it will only modify the resulting x as a second-order effect. The important driver for segregation is the y component, which we now denote simply as v. In the absence of segregation, $v=0$, the particles would simply be 'passively' advected by the mean flow. The segregation process is the consequence of the dependence of the effective advection flow on the particle characteristic, which we will refer to as 'non-passive' advection.

There are basically two essential mechanisms which can be invoked for justifying the non-passive advection of the small particles.

- The first one, referred to as a 'convective' process, can be modeled as a 'percolation' process. We will argue that this mechanism is the dominant one at play in the experiment, and the next section will concentrate on its modeling. Before this, we would like to discuss another possible mechanism which is believed to be much weaker.
- The second mechanism focuses on the diffusive-type motion of the particles due to collisions. The segregation occurs there in a very different way: a small particle in a large particle environment will have a large diffusion coefficient since its random motion is counted in units of the large particle diameter, whereas, in a sea of small particles, the diffusion will be drastically reduced. The driving mechanism for segregation is thus the relative change in diffusion coefficient due to the local concentration of particles. In this case, however, one should rather focus on large particles. Indeed, the limiting process will then be to expel large particles from the central core, because their diffusion coefficient is naturally much smaller than that of the smaller disks, and even more so if they are trapped in the central core. This contradicts the observation, since large particles are easily extracted from the central core in the first cycle. Moreover, this differential diffusion, is a cause of '*intrinsic segregation*' rather than '*structural segregation*'. By this we mean that small particles should tend to gather in clusters, but anywhere in the flow as observed experimentally, e.g. on a blowing table [15], and nothing will favor a central core close to the axis of rotation. The convection is, on the other hand, an example of a structural segregation since the large structure scale will dominate

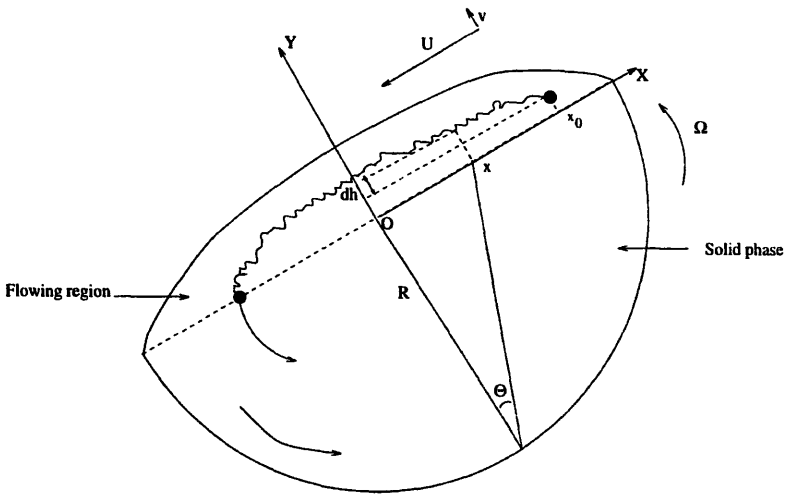

Fig. 5. Sketch of the small particle trajectory into the flowing phase.

local fluctuations. We thus believe that the convective mechanism is a more realistic description of the process. Let us stress however that if diffusion alone seems difficult to advocate, it is certainly an effect which may assist the convection mechanism.

7. Differential convection

In the convective case, v is a mean systematic drift which results from the larger mobility of the small particles. In the flow, a small particle can fall into an opening gap between two large particles, whereas a larger one could not by steric hindrance. Thus, in a sheared layer, a small particle is expected to have a downward velocity larger than that of big particles. The mobile large particles act as a fluctuating sieve, letting small ones moving downward squeeze in easily and then gain an additional downward velocity.

The drift v is proportional to the rate at which a gap may open. One potential downward motion will happen after a fixed strain of order 1, so that the small particle moves to a neighboring disk. The mean y displacement is equal to the large disk size times the probability that a gap opens. Without specifying the latter probability, we may simply state that v is proportional to the shear rate γ [16]:

$$v = A\gamma$$

where A depends on the sizes of the particles, and on the surrounding concentration of small or large disks. A is difficult to estimate, although its role is obviously major in the segregation. However, in the dilute approximation, we do not have to specify the efficiency of the segregation mechanism with the local environment. It suffices to say that A is zero in the region of the flow which consists of small particles, and assumes a constant value everywhere else in the flowing region.

Let us now imagine a small particle starting at x_0 (see Fig. 5). We want to compute the probability distribution of x_1 at which it will be captured by the rigid cluster. We first discard the diffusion due to collisions and consider only the mean systematic drift v. The small disk enters in the flowing region at a distance $h(x_0)$ from the free surface. In the flow, this distance increases to $h(x_0)+\mathrm{d}h$ due to the transverse velocity, so that $\mathrm{d}h = vt(x_0)$, where $t(x_0)$ is the time needed to flow from x to $-x$ by passive advection:

$$t(x_0) = \int_{-x_0}^{x_0} \frac{1}{\gamma(h(x') - h(x_0))}\,\mathrm{d}x'$$

$$= \frac{1}{\gamma\eta} \int_{-\theta_0}^{\theta_0} \frac{\cos(\theta)\,\mathrm{d}\theta}{\cos(\theta) - \cos(\theta_0)} \qquad (9)$$

where $\theta_0 = \arcsin(x_0/R)$. The above integral is logarithmically divergent for $x \approx \pm x_0$ which is clearly not physically realistic. This weak divergence can be removed by introducing a finite cut-off in the velocity, say at the scale of the disk below which a continuum breaks down. The final result is simply that to the leading order $t(x_0) = t_0$ is independent of x_0 and so is $\mathrm{d}h = vt_0$. At the end of the flow period, the outlet x value amounts to $x_1 = x_0 - \mathrm{d}x$, so that $h(x_1) = h(x_0) + \mathrm{d}h$, and hence:

$$x_1 = x_0 + vt_0/h'(x_0) \qquad (10)$$

where h' is the derivative of h. We finally obtain

$$x_1 = x_0 + \frac{A\gamma t_0}{\eta}\frac{(R^2 - x_0^2)^{1/2}}{x_0} \qquad (11)$$

This recursion relation is only valid for a large x_0 where h' is large enough so that $\mathrm{d}x$ remains small. As x_0 approaches zero, the change in height cannot be considered as constant any longer. Again a finite cut-off has to be introduced at the scale of the disk size.

It is interesting now to exploit the recursion relation on x. We observe that as x_0 approaches R, $x_1 - x_0$ goes to zero. Thus, the outer boundary appears to be a fixed point in spite of the tendency of the small particles to flow across the y direction. Obviously, this fixed point is unstable, but the time

needed to escape from it may be long. Starting from $x_0 = R - \epsilon$, it is a simple matter to show that, after n cycles, x_n scales as

$$R - x_n \propto \left(\frac{A\gamma t_0}{\eta R^{1/2}}\right) \epsilon n^{1/2} \tag{12}$$

provided that $\epsilon n^{1/2} \ll R$. Therefore, the number of cycles n^* needed to escape from the outer boundary of the drum has a scaling which can be obtained simply. We require that $R - x_n$ is a fixed fraction of R, and ϵ is of the order of the small particle size. Using the expression $\eta \sim (\Omega/\gamma)^{1/2}$ cf. Eq. (7), we obtain the dependence of n^* on Ω as

$$n^* \propto \Omega \tag{13}$$

When expressed in real time, we get the time needed to escape from the boundary as $\tau \propto n^*/\Omega$, i.e. a time which does not depend on the rotation velocity! This conclusion, experimentally verified (see Fig. 3), is surprisingly simple, although it results from a subtle balance between the increase of cycles due to the deeper flowing region and a decrease simply due to the rotation velocity. Finally, we note that the computed escape time is the largest one encountered in the flow, so that it directly gives the segregation time. Moreover, as this process is localized in space at the outer boundary, the size of the core region does not affect it. Therefore, we predict that the segregation time is independent of the concentration in small particles.

8. Experimentally determined transition probabilities

In the previous sections, we had to resort to a number of hypotheses in order to reach definite conclusions. It is possible to measure directly the transition operator $p(x|x')$ experimentally from the image analysis technique. This section shows how to compute directly the segregation time from the experimentally determined transition probabilities.

8.1. Experimental analysis

We have studied the trajectories of only one (small or large) particle in a mixture and shown that, to first order, the segregation can be regarded as a single-particle process [12–14]. Therefore, in this study, we have carried out experiments with only one small tracer in the mixture distinct from the other (black) particles by painting it in white. The experiment is filmed for a duration of 20 min. We can follow the small white disk and record its coordinate, x, just before it is injected in the flow and just after it is trapped into the solid phase. We discretize the x variable in 11 intervals of length equal to two large disk diameters (chosen so as to limit the noise in the data acquisition). The discretized values of p and f are respectively the coefficients of the matrices $\mathbf{T}$ and $\mathbf{F}$. We estimate the coefficients of $\mathbf{T}$, $t_{i,j}$, from 1000 pairs (x', x) of the tracer on the liquid phase:

$$t_{i,j} = p(x_i|x_j) \tag{14}$$

The normalization conditions are written for all j:

$$\sum_{i=1}^{n} t_{i,j} = 1 \tag{15}$$

We compute the 11 eigenvalues of $\mathbf{T}$ which are real or complex conjugates and we classify the eigenvalues in decreasing order of their modulus. We note λ_i, the ith eigenvalue, and (Φ_i) the associated eigenvector.

8.2. Asymptotic steady state

Let $\mathbf{F}^\infty$ be the one-dimensional matrix $\mathbf{F}$ in the stationary state. Eq. (5) gives

$$\mathbf{F}^\infty = \mathbf{T}\mathbf{F}^\infty \tag{16}$$

So that $\mathbf{F}^{(\infty)}$ is an eigenvector of $\mathbf{T}$, with eigenvalue unity. Since no eigenvalue can exceed unity, $\mathbf{F}^{(\infty)}$ is identical to Φ_1. The calculation of the coordinates of $\mathbf{F}^{(\infty)}$ gives

$$\mathbf{F}^{(\infty)} = \Phi_1 = \begin{pmatrix} 0.02 \\ 0.05 \\ 0.10 \\ 0.13 \\ 0.11 \\ 0.14 \\ 0.11 \\ 0.10 \\ 0.07 \\ 0.08 \\ 0.08 \end{pmatrix} \tag{17}$$

It should be emphasized that the transition probabilities are computed with a uniform measure on x. However, the latter does not correspond to a uniform measure in area for the drum. Hence, to express the matrix $\mathbf{F}$ in physical units, one may resort to the concentration of small particles in shells $x_i \le x \le x_{i+1}$. The latter is a density vector, denoted $\boldsymbol{D}$. From $\mathbf{F}$ we can calculate the density vector, $\boldsymbol{D}$, of small particles in the different shells, whose surface is called S_i. The value of area S_i is

$$S_i = \frac{1}{2}\pi(r_i^2 - r_{i-1}^2) \tag{18}$$

where r_i, as shown in Fig. 6, is the radius of the half-circle joining the two symmetrical values of x and x'. We deduce

$$D_i = \frac{F_i}{S_i} \tag{19}$$

The uni-dimensional matrix $\mathbf{D}^\infty$ represents the probability of the presence of small disks in the steady state, for an extreme dilution (one small disk). We plot in Fig. 7 the value of $\mathbf{D}^\infty$ versus x'. It should be noted that already a clear segregation appears. This is a direct proof of this phenomenon being a structural instability, which may be studied as a sin-

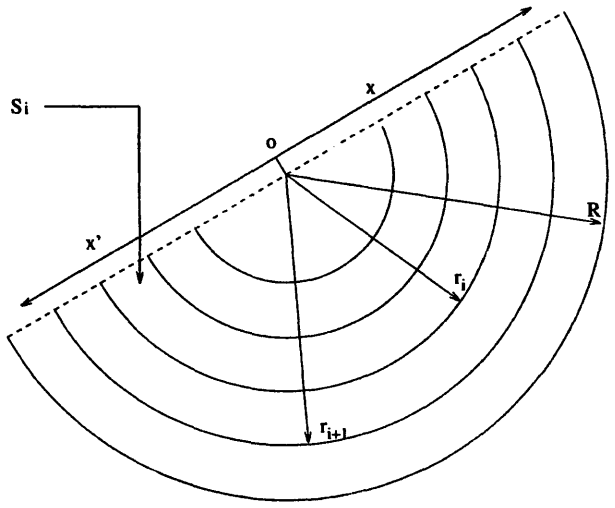

Fig. 6. Sketch of the discretization of the drum into 'shells'.

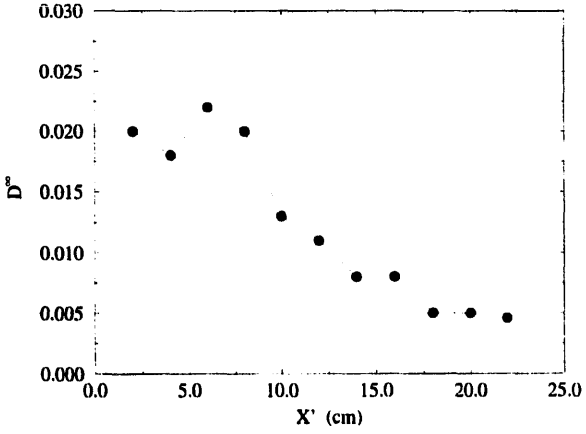

Fig. 7. Asymptotic density of small particles from the drum axis estimated by the steady-state eigenvector of the transition operator for an extreme dilution.

gle-particle process (not a collective one), at least in the dilute regime. This confirms the previous theoretical analysis. It also shows the limitations of our approach: the segregation is not complete. The small particle can freely move inside a core of size roughly equal to 8 cm. This suggests the relevance of diffusion in the complete description of the flow. It might however not be a crucial point in the determination of a segregation time since we have argued that the escape from the outer boundary was the limiting process.

8.3. Segregation time

In order to compute the order parameter, we introduce the indicator of the reference cluster, $\mathscr{C}$, such that $c_i = 1$ if $x_i \in \mathscr{C}$. The normalized surface after k periods (semi-revolutions), called $a(k)$ can be written as

$$a(k) \equiv \mathscr{C}^t \mathbf{F}^{(k)} \tag{20}$$

where $\mathscr{C}^t$ is the transposed matrix of $\mathscr{C}$. The order parameter assumes the following expression:

$$P_o(k) = \frac{\mathscr{C}^t(\mathbf{F}^{(k)} - \mathbf{F}^{(0)})}{\mathscr{C}^t(\mathbf{F}^{(\infty)} - \mathbf{F}^{(0)})} \tag{21}$$

or

$$P_o(k) = \frac{\mathscr{C}^t(\mathbf{T}^k - \mathbf{I})\mathbf{F}^{(0)}}{\mathscr{C}^t(\mathbf{T}^\infty - \mathbf{I})\mathbf{F}^{(0)}} \tag{22}$$

where $\mathbf{I}$ is the identity matrix. Eq. (22) becomes, in the eigenvector basis, writing $F^{(0)} = \sum \alpha_i^{(0)} \boldsymbol{\Phi}_i$:

$$P_o(k) = \frac{\sum_{i=1}^{n} (\lambda_i^k - 1)(\mathscr{C}^t \boldsymbol{\Phi}_i)\alpha_i^{(0)}}{\sum_{i=1}^{n} (\lambda_i^\infty - 1)(\mathscr{C}^t \boldsymbol{\Phi}_i)\alpha_i^{(0)}} \tag{23}$$

We know that λ_1 is equal to unity (see Section 4.1). For $i > 2$, the modulus of the eigenvalues is smaller than unity, so λ_i^∞ tends towards zero. Then Eq. (23) becomes

$$P_o(k) = -\sum_{i=2}^{n} (\lambda_i^k - 1)\beta_i \tag{24}$$

where

$$\beta_i = \frac{(\mathscr{C}^t \boldsymbol{\Phi}_i)\alpha_i^{(0)}}{\sum_{i=2}^{n} (\mathscr{C}^t \boldsymbol{\Phi}_i)\alpha_i^{(0)}} \tag{25}$$

and

$$1 - P_o(k) = \sum_{i=2}^{n} (\lambda_i^k)\beta_i \tag{26}$$

$$1 - P_o(k) = \sum_{i=2}^{n} \beta_i \exp[-k(\ln(\lambda_i))] \tag{27}$$

The latter equation allows us to re-interpret the λ_i in terms of relaxation time τ_i such that

$$\tau_i = \frac{-1}{\ln|\lambda_i|} \frac{\pi}{\Omega} \tag{28}$$

Let us go back to the experimental results. As previously mentioned in Section 3.2 we showed experimentally that the order parameter follows an exponential law with time. We have

$$P_o(t) = 1 - \exp\left(\frac{-t}{\tau^*}\right) \tag{29}$$

We denote τ^* the experimental time constant in order to distinguish it from the value of τ_i used in the calculation. The experimental value of τ describes the system behavior in the long time limit, and thus it is identified with the maximum of the τ_i set, i.e. with τ_2. So we can identify from Eqs. (25) and (26):

$$\tau^* = \tau_2 = \frac{-\pi}{\Omega \ln|\lambda_2|} \tag{30}$$

where λ_2 is the largest eigenvalue different from unity.

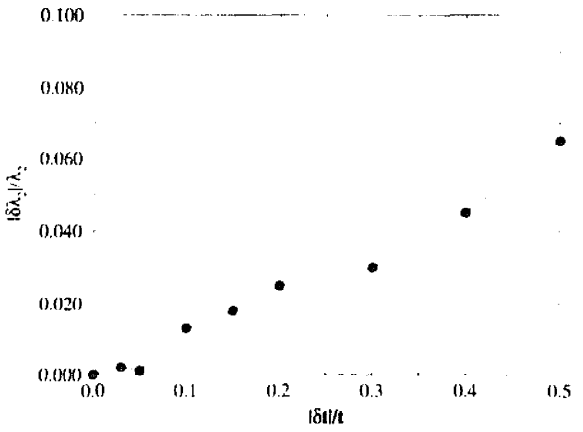

Fig. 8. Variations of the second largest eigenvalue of the transition matrix, $|\delta\lambda_2|/\lambda_2$ as a function of the imposed perturbation on **T**, $|\delta t|/t$.

8.4. Results and discussion

Let us go back now to the study of the matrix **T**. The value of λ_2 is equal to 0.44. For $i > 2$, the modulus of the eigenvalues is very small, smaller than 5×10^{-2}. This explains that only the dominant τ is observed experimentally and not the full series written in Eq. (25). From Eq. (27), the value of τ is equal to 26 s, a value close to the experimental one, which is equal to (29 ± 6) s (see Section 3.2).

The determination of the error bars on our estimate of τ from the measured **T** is a difficult point. A direct measurement of **T** requires about 1000 recorded pairs (x,x'). Thus, we propose to check numerically the stability of the second eigenvalue λ_2 from which τ is estimated. Therefore, we have tried to alter randomly the stability of the coefficients $t_{i,j}$ under some specific constraints, and check the fluctuations of λ_2.

The perturbation of $t_{i,j}$ is done while preserving the normalization condition $\sum_i t_{i,j} = 1$. We also imposed naturally that these transition probabilities remain positive. Finally, we choose a variable amplitude of perturbation $|\delta t|/t$. We show in Fig. 8 the value of $|\delta\lambda_2|/\lambda_2$ (λ_2 being the only one eigenvalue value of interest in this calculation) versus $|\delta t|/t$, the perturbation of the largest **T** coefficient. As shown in Fig. 8, the value of λ_2 is very stable when $|\delta t|/t \leq 0.3$. We also note that the other eigenvalues are very stable too. This result is important because it shows that the value of τ obtained from this calculation remains stable in spite of the parameter fluctuations present in our determination of **T**.

9. Conclusions

In this paper, we have analyzed the radial segregation in a two-dimensional rotating drum.

This segregation, which is a very fast process, has been found to be experimentally independent of the concentration of small particles and of the rotation speed Ω.

Starting from the experiment, we have proposed a theoretical analysis of this phenomenon, whose origin takes place in surface flow. The segregation is then described as being a single-particle process, which is essentially controlled by a differential convection of the small particles in the flow. By simply stating that the relative motion velocity, v, of small particles in the 'mean flow' is proportional to the shear rate γ, we have shown that the segregation time is independent of the rotation velocity — a surprising result but one which we have experimentally verified — and of the concentration of small particles.

We finally use the results obtained in our experimental analysis of the surface flow to write the transition matrix, **T**, whose coefficients are the conditional probabilities $p(x'|x)$, giving the probability that a particle which reaches the flow at a position x is trapped in the position x'. We are then able to estimate the time constant of the process from an experimental determination of these conditional probabilities.

In this analysis, essentially done in the dilute regime, the surrounding concentration of small and large particles has been considered as irrelevant to the segregation process. As we have already described in the Introduction, two kinds of segregation appear in the three-dimensional device, and the process coming to the axial segregation may be different from the mechanism we analyzed in the two-dimensional case. Some recent works [11,17] claim that the axial segregation is explained by a diffusion process generated by different values of the dynamic angle of repose according to the particle size.

10. List of symbols

$a(t)$	normalized surface occupied by small particles in the reference cluster, at time t
c_f	flowing bulk density
c_s	bulk density in the solid region
C	concentration of small particles
$\mathcal{C}$	reference cluster
$\boldsymbol{D}$	density vector
$f_n(x)$	distribution of x values after n cycles
$f_x(x)$	radial distribution of small particles
$f_0(x)$	initial distribution of x values for small particles
F	matrix of components $f(x_i)$
$h(x)$	thickness of the flowing zone
I	identity matrix
$p(x'\|x)$	conditional probability
$p(x\|y,C)$	conditional probability at concentration C
$p^{(l)}$	conditional probability for a large particle
$p^{(s)}$	conditional probability for a small particle
P_o	order parameter
R	drum radius
S_i	surface between the shelves of radius x_i and x_{i+1}
T	matrix of components $t_{i,j}$ with $t_{i,j} = p(x_i\|x_j)$
U	velocity of the large particles in the flowing region (Section 6)

v	transverse (along y) component of the small particles velocity
x_c	radius of the central segregation core
x_i	shell index referring to the radial discretization
y	coordinate normal to the free surface

Greek letters

γ	shear rate in the flowing phase
λ_i	ith eigenvalue of $\mathbf{T}$
τ	time constant
$\Phi(x)$	flow in the flowing region
Φ_i	eigenvector associated with λ_i
Ω	rotation speed of the drum

Acknowledgements

It is a great pleasure to thank Eric Clément, Jacques Duran and Jean Rajchenbach for numerous fruitful discussions, which have guided all the experimental work. This work was partly supported by a grant from MRT (Contract No. 92T0143). We also acknowledge the support of the CNRS Groupement de Recherche 'Physique des Milieux Hétérogènes Complexes'.

References

[1] A. Rosato, F. Prinz, K.J. Strandburg and R.H. Swendsen, Powder Technol., 49 (1986) 59.

[2] A. Rosato, K.J. Strandburg, F. Prinz and R.H. Swendsen, Why the brazil nuts are on top: size segregation of particulate matter by shaking, Phys. Rev. Lett., 58 (1987) 1038.

[3] R. Jullien and P. Meakin, A mechanism for particle size segregation in three dimensions, Nature, 344 (1990) 425.

[4] R. Jullien and P. Meakin, Three-dimensional model for particle-size segregation by shaking, Phys. Rev. Lett., 69 (1992) 640.

[5] J. Duran, J. Rajchenbach and E. Clément, Arching effect model for particle size segregation, Phys. Rev. Lett., 70 (1993) 2431.

[6] S.B. Savage and K.L. Lun, Particle size segregation in inclined chute flow of dry cohesionless granular solids, J. Fluid Mech., 5189 (1988) 311.

[7] S.B. Savage, Flows of Granular Materials with Applications to Geophysical Problems, A Course of Lectures, 1992.

[8] T.R.H. Davies, Large debris flows: a macro-viscous phenomenon, Acta Mech., 63 (1986) 161.

[9] T.H. Erismann, Flowing, rolling, bouncing, sliding: synopsis of basic mechanisms, Acta Mech., 64 (1986) 101.

[10] M. Nakagawa, S.A. Altobelli, A. Caprihan, E. Fukushima and E.-K. Jeong, Non-invasive measurements of granular flows by magnetic resonance imaging, Exp. Fluids, 16 (1993) 54.

[11] O. Zik, D. Levine, S.G. Shtrikman and J. Stavans, Phys. Rev. Lett., 73 (1994) 5.

[12] F. Cantelaube and D. Bideau, Radial segregation in a 2d drum: an experimental analysis, Europhys. Lett., 30 (1995) 133–138.

[13] F. Cantelaube, Ecoulements granulaires dans un tambour à deux dimensions, segregation, Thesis, Rennes, 1995.

[14] E. Clement, J. Rajchenbach and J. Duran, Mixing of granular materials in a bidimensional rotating drum, Europhys. Lett., 30 (1995) 7–12.

[15] C. Annic, Milieux granulaires 2d: distribution de taille, segregation, Thesis, Rennes, 1994.

[16] S.B. Savage and K.K. Lun, J. Fluid. Mech., 189 (1988) 311.

[17] D. Bideau and A. Hansen, Disorder and Granular Media, North-Holland, Amsterdam, 1993.