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Evidence of two effects in the size segregation process in a dry granular media

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In a half-filled rotating drum, the *size segregation of particles of equal density* builds a ring pattern of the large particles, whose radius continuously varies from the periphery to the center depending on the size ratio between particles [Thomas, Phys. Rev. E 62, 1 (2000) 961-974]. The existence of these circles with an intermediate radius shows that the segregation at an intermediate level within a flow is possible. It is interpreted as a consequence of a balance, at that level, between two segregation effects: a mass effect and a geometrical effect, both due to the size difference. Because the difference of mass can be due to a size difference, but also to a density difference, in this paper, we experimentally vary the mass effect by studying the segregation of particles of *different densities* and sizes in a half-filled rotating drum and other devices (chute flow, pile). In the drum, the radius of the ring continuously varies from the periphery to the center and is very sensitive to both the size and density ratios compared to characteristics of the small beads of the bed (from 0.36 to 4.8 for the density ratio, from 1 to 33 for the size ratio). For the densest beads, the circle is close to the center, for the lightest beads it is close to the periphery. The excess of the mass (due only to a size excess, a density excess, or both) leads heavy beads to segregate deep inside, by a push-away process. The excess of the size leads large beads to segregate at the surface, by a geometrical dynamical sieving process. The radius of the segregation ring is determined by the relative intensities of both effects. For example, for particles of equal density, the geometrical effect is dominant for small size ratios (typically <5): large beads are at the surface; and this effect is negligible for high size ratios (typically >15): large beads are close to the bottom. Using the fact that the ring formation corresponds to a balance between these two effects, we can study the geometrical effect, and thus the size segregation, by using the whole set of data acquired on the locations of segregated beads of different densities and sizes.

PACS numbers: 45.70.Mg, 45.70.Cc, 45.70.Ht

I. INTRODUCTION

When a granular matter composed of several types of particles is put into motion, there is a de-mixing of the different types of particles. This segregation appears whenever there are differences in size, density, shape, roughness... For an example, in a initial mixture of large and small beads composed of the same matter, the large beads are often found at the surface of the bed after the granular matter has been put into motion [1] or vibrated [2], or at the periphery of a rotating half-filled drum [3, 4, 5]. Segregation of beads of different sizes has been interpreted as a dynamical sieving of particles during the flow. The sieving allows the small particles to go down, towards the bottom and, consequently, the large ones end up at the surface. This process comes from the size difference and is actually a geometrical effect. However, this interpretation is incomplete because it does not take into account the fact that large beads may push away their neighbors to go downwards [6, 7], and can get buried into the bed. In a previous paper [6],

we investigated the balance between the two effects implied in the size segregation, named the geometrical and mass effects: the geometrical sieving pushes the large bead to the surface of the granular matter, as the mass effect pushes the large (and heavy) beads to the bottom. In three experimental devices (flow in an inclined channel, formation of a heap, rotation in a half-filled drum) a mixture of large and small beads of the same density has been sheared during the flow. For a small fraction of large beads (typically 3%), the location of the large beads continuously evolves (intermediate segregation) from the surface (classical figure of segregation) to the bottom (reverse segregation), when increasing the size ratio. Typical values are: for size ratios between 1 and 5, large beads are still visible at the surface, and for size ratios above 15, they are very close to the bottom. This evolution is masked when using a large fraction of large beads (for example 50%). We concluded that for small size ratios the geometrical sieving is dominant and determines the location of the beads to be at the surface of the bed. For larger particles, the mass effect is dominant and the large beads are found in a reverse location, i.e. buried near the bottom of the flowing zone. But for intermediate size ratios, the large beads are found at a particular level inside the bed, whose location depends on the value of the size ratio. This implies that these 2 effects exactly counterbalance at this level in the flow, from which we

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can deduce that at least one of the two effects depends on the vertical position within the flow.

This intermediate segregation produces rings of segregated tracers when the mixture is placed in a rotating drum [Fig. 1]. The radius of the ring can be accurately measured, and is very sensitive to the size ratio between tracers and beads of the bed. The location of the ring is then closely related to the relative intensities of the geometrical and mass segregation effects. When the geometrical effect is the strongest, the ring is close to the periphery, and when this effect is the smallest, the ring gets closer to the center. In most previous studies, it is only possible to know if segregated particles are at the surface or at the bottom of the flow, i.e. there are only two possible patterns of segregation (or three when counting also the homogeneous state). But contrary to these classical patterns of segregation, the evolution of the location of the ring is continuous: there is a infinite number of patterns. Consequently, it is possible to continuously quantify the relative intensity of the segregation effects by the measurement of the radius of the ring.

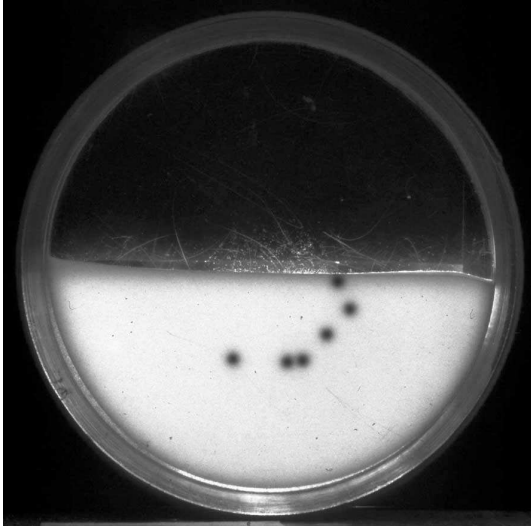


FIG. 1: Ring of segregation of 1.5-mm large tracer beads (Glass) in a bed of 90- μm small Glass beads. The radial location of tracers is very precisely defined (note that the upper bead is still in the flowing layer). The location is very sensitive to the value of the size ratio [6] and of the density ratio [this paper]. The variation of the radius of the ring is a powerful means to study the geometrical effect of segregation by making an equivalence with the variation of radius induced by a change of the density ratio.

A variation of the radius of the ring can be due to a change of the size ratio (changes of both mass and geometrical effects), but also of other parameters, such as the density ratio (only change of mass effect). Consequently, the existence of these well-defined rings whose radius continuously varies provides a way to make a quantitative link between the unknown intensity of the geometrical segregation effect and the intensity of another

segregation effect (here, the mass effect).

The aims of the present study are (1) to confirm the interpretation of a mass effect as being responsible of the reverse and intermediate positions of large beads of equal density after size segregation [6] and (2) to obtain data to quantitatively separate the two effects (geometrical and mass) implied in the process of size segregation of beads of equal density. For that reason, we vary the mass effect independently of the geometrical effect, by changing the density of the tracer. We then experimentally investigate the segregation of beads of different densities, for each size ratio.

Experimentally, the segregation due to a density difference has been less studied than the segregation due to a size difference, one of the reasons being probably the difficulty of obtaining beads of various densities. Some studies focus on the phenomena happening during the shaking of a container filled with a mixture of granular matter [8, 9] or follow the trajectory of tracers during the vibration [10, 11]. The previous experimental studies, which involve a granular matter sheared into motion in a slow frictional way report that for equivalent sizes, the heavy particles are found at the bottom of the bed (or center of the drum), and that the light particles are at the surface (or periphery of the drum) [12, 13, 14]. But they mostly concern large density ratios (from 3 to 7). Numerical studies have also been done [13, 14, 15, 16, 17] using a large and continuous range of density variation. Some results are close to a ring segregation and show a continuous evolution of the final location of the tracers versus the mass ratio [17]. When using particles of different sizes and densities, two effects are in addition or in competition, leading to up, down or no segregation [18, 19]. For example, a difference of size (ratio 3) can dominate a small difference of density (ratio 1.5), leading to large heavy particles on the surface [20]. Experimentally [19, 21] or numerically [18], adjusting density and sizes can lead to a homogeneous bed which has been interpreted as a global balance between the effects of density and size. But these studies concern only the segregation state (homogeneous or not) of the whole granular matter because they use similar fractions of large and small particles, contrary to the segregation pattern in rings which gives both the segregation intensity (ranging from homogeneous to totally segregated) *and the location* of the segregated minority particles.

The first and main part of the present study focuses on the ring segregation pattern in a drum. It involves beads with small to large size ratios (1 to 33), and different density ratios (from 0.36 to 4.8). We are interested in the problem of one single large tracer in a bed composed of identical small beads, and we get data on its final segregated location within the flow (corresponding to the radius of the ring). The aim is to relate this location to the forces that have been exerted on this tracer for it to go and stay at that particular equilibrium level during the flow. In that way, we can study the forces responsible for the segregation, knowing (1) the location

where they balance, and (2) how this varies with the size and density ratios. We then choose to work with very few large beads in a bed of small ones: volume fractions are from 0.16% to 3%. Consequently, there is no percolation process, even for large size ratios. The fractions are low enough such that the large beads are considered as individual tracers, but tracers are numerous enough to allow some statistics on the data (10 to 50 beads). These experiments might not be called “experiments of segregation between two species” as there is no de-mixing when considering the extreme case of one single large bead in the bed of small ones. In that point of view, the segregation process is considered as being mainly a one-particle process as shown by [3, 4] and is the result of all large identical beads having the same preferential location in the bed: they all go to this location, and consequently regroup.

We are aware of the fact that interactions between large beads could modify the process but do not want to treat that point in this article. Briefly, it has been experimentally observed that the final location of a species depends on its volume fraction in the mixture [6]: at high fractions of large beads, their average location is shifted towards the surface. The interpretation involves the reduction of the push-away process due to the presence of other large and heavy beads in the medium. The results of this paper therefore cannot be accurately extended to the case of a high fraction of large beads, even if most of the features are still qualitatively valid.

Our study concerns only experimental devices where particles are submitted to a frictional shear motion during the flow (no vibration, no collisional flow). Two sets of experiments have been done. In the first part, we precisely measured in a half-filled rotating drum, the locations of few large tracer beads of different densities, placed in a bed of small Glass beads whose size has been varied. In the second part, experiments have been done in three experimental devices to show that the reverse and intermediate segregations are not only characteristic of the drum device: formation of a pile, half-filled rotating quasi 2D drum, and chute flow along a slope have been used. Various mixtures of particles and beads have been done (with size ratios from 0.2 to 10, and density ratios from 0.5 to 3.2), and only the general behavior of the segregation (up-inside-homogeneous) have been determined as a function of both density and size ratios. A link between these two parts is made through values of intermediate mean locations of tracers.

II. EXPERIMENTS IN A ROTATING DRUM

The segregation in a half-filled drum has been experimentally studied because the radial segregation is easily quantified in such a system [3, 4, 6]. The drum turns around its axis, which is placed horizontally. An initially homogeneous mixture of two types of beads flows in a

slow frictional regime. The flowing layer is located on the free surface. The remainder of the beads rotates in a solid way with the drum rotation. Two de-mixing processes take place in a drum: radial and axial segregations [22, 23, 24]. The axial segregation takes longer to appear than the radial segregation. Here, we study the radial segregation, and avoid the axial segregation by rotating only a few revolutions.

We follow the behavior of the minority component (indexed 2, and named the tracers) in a bed made of the majority component (small Glass beads, indexed 1). A mixture is characterized by the diameter d_2/d_1 and the density ρ_2/ρ_1 ratios. We limit the drum study to the case of large tracers, i.e. $d_2/d_1 \geq 1$.

We quantify the segregation by measuring the distances r_i from the center of tracer beads to the center of the drum [Fig. 2]. r is the average of r_i , and R is the radius of the cylinder. Two parameters are involved: (1) the spreading of the r_i distribution is related to the intensity of the segregation. When the standard deviation of the r_i distribution is low, the intensity is strong, and the segregation pattern is well focused. When the standard deviation is large, the segregation is weak and tracers are widely spread around their mean location. (2) The mean value of r_i indicates the location of the segregated tracers whether the segregation is strong or weak. It is possible to relate this radius to the location within the flow. In fact, each radius r_i corresponds to a ‘vertical’ level z_i in the flowing layer (‘vertical’ meaning along an axis perpendicular to the flow surface) [25]. Roughly $r_i=0$ comes from a tracer whose center is at the bottom of the flow, and $r_i=R$ from a tracer whose center is at the surface (half-embedded). The link is done by the shape of the interface between static and flowing zones, which can be approximated by a parabolic function when the free surface is plane [26, 27]. More precisely, the large bead may stop when its bottom touches the interface between the flowing layer and the static part. In that case, r_i would be smaller than the radius corresponding to the intersection between z_i and this interface. That explains why beads whose centers are above the free surface present larger r_i than beads just half-embedded, although a simple intersection linking z_i and r_i would give a maximal value for r_i for all beads which centers are above the surface, whatever z_i is. This remark is found on qualitative observations, and we do not have data to establish a link between z_i and r_i . We assume that r_i and z_i are linked by a parabolic function, as if they were determined by the shape of the interface, but the multiplicative constant may be different from that corresponding to the interface curve.

A. Experimental device

The drum is 42 mm long and 48.5 mm in diameter, made of Glass and Steel (caps), and half-filled with the homogeneous mixture of small Glass beads and large

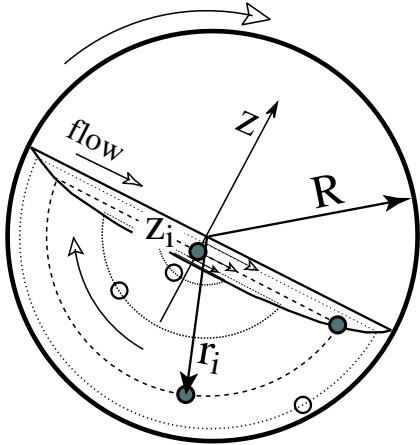


FIG. 2: Each measured radial position r_i in the rotating solid part corresponds to a perpendicular ‘vertical’ location z_i in the flowing layer.

TABLE I: Beads used in the drum experiments

Materials :	density (g/cm ³)	sizes (mm)
Polypropylene (PP)	0.9	3
Polyamide (PA)	1.14	3
Polyacetal (POM)	1.41	3
Teflon (PTFE)	2.15	3
Glass*	2.5	3-3.14
Aluminum (2 sizes)	2.7	3.97; 2.5
Silicone Nitride (Si ₃ N ₄)	3.16	3.175
Ceramic (Al ₂ O ₃)	3.9	3
Titane	4.52	3
Zircone (ZrO ₂)	6.0	2
Steel (3 sizes)	7.85	1; 2.5; 3
Lead*	12	3.02-3.07
*not perfectly spherical		

tracer beads. The large tracer beads are usually 3 mm in diameter, have different densities (table I) and represent from 0.16% to 3% of the volume of the beads. We consider that they do not interact and do not modify enough the characteristics of the medium for their location to be affected [6]. The variation of the density ratio has been obtained by changing the density of the tracers, the variation of the size ratio by changing the size of the small Glass beads. Small Glass beads are very-well sorted beads (less than 10% of relative spreading around the mean value [6]) in order to work with “mono-disperse” size populations. Mean diameters of the ranges of Glass beads go from 90 microns to 4 mm: 90; 150; 180; 200; 212; 250; 300; 355; 400; 425; 500; 600; 710; 850; 1000; 1500; 2000; 2500; 3000; 4000 μm . The relative density of the Glass is 2.5 (measured with a picnometer from 2.46 to 2.54 with no systematic variation with the size).

In order to minimize electrostatic and humidity cohesive effects between beads, the humidity in the laboratory room is held between 50-55% [28] and special care

is given to the experiments involving the smallest beads. Electrostatic effects are nevertheless strong when using Teflon beads which attract small Glass beads.

The rotation is manually obtained by rolling the drum on a plane for a third of the experiments (with no systematic order compared to size or density values). The last experiments were performed with a motor, and do not show any difference. Rotation speeds are chosen to correspond to a continuous flow on the free surface of the bed and to a plane free surface. They are usually around 0.085 s^{-1} (from 0.04 to 0.1 s^{-1} , up to 0.15 s^{-1} in few cases written on the figure captions). But experiments with the largest Glass beads for the bed (2-2.5-3-4 mm) have been systematically done with higher speeds to obtain a continuous flow (respectively 0.1 - 0.12 - 0.13 - 0.2 s^{-1}).

After a few revolutions (3 to 6), we carefully dip the drum into water. It is then possible to open it without disturbing the spatial distribution of the beads. For the largest Glass beads, the drum is frozen before opening it. Several cross-sections are carefully made perpendicular to the axis of the drum through all the deposit, to measure the radial location r_i of each tracer [Fig. 2]. Roughly, r_i/R varies from 1 (tracers segregated at the surface of the flow) to 0 (tracers at the bottom of the flow). More precisely, and because of the finite size of the tracers, the maximum value of r_i/R is equal to $(1-d_2/2R)$ (0.938 for 3-mm tracers), corresponding to the case where the large bead is touching the periphery. Because of the thickness of the flowing layer, the minimum value of r_i/R is not equal to 0 either. As the densest tracers are not visible during the flow, whatever the size of the small bead used, we assume that the thickness of the flowing layer is at least as big as a large bead: d_2 . For the measurements we do not take into account the beads located at less than $1.5d_2$ from the free surface, because they could be touching the flowing layer. Due to this thickness, the value of r_i/R is at least superior to $1.5d_2/R$ (0.18 for 3-mm tracers). We noticed that the tracers touching the caps have a slightly different mean location from tracers inside the bed, and removed them for the measurements.

B. Circles of segregation

After segregation, the large tracer beads are found on a half-circle inside the drum, as it has been previously observed [6]. The radius of this circle varies with the characteristics of the beads of the mixture [Fig. 3]. After one turn, the radius of the ring is constant when increasing the number of revolutions [6] corresponding to the stable pattern of segregation obtained after one passage in the flowing layer [29]. The convergence towards this stable pattern is more rapid than in the case of a 2D drum [5]. Nevertheless, measurements of convergence have been done only for a size ratio 6, and consequently the drum is rotated 6 turns in the case of the highest speeds (lowest size ratios), to take into account a poten-

tial increase of the time of convergence to a final segregated state [5].

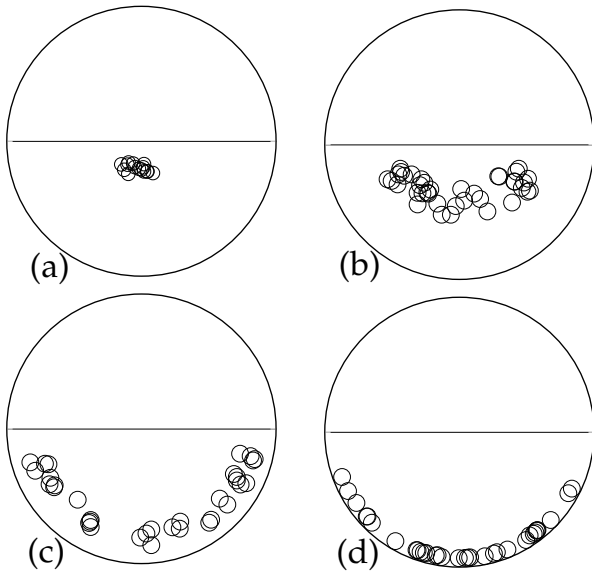


FIG. 3: Some examples of the locations of the tracers in a bed of small Glass beads. Several cross-sections from one single experiment have been added on each drawing. Material of tracer & diameter of small Glass beads are: (a) Steel (2.5 mm) & 300 μm , (b) Al_2O_3 (3 mm) & 710 μm , (c) Teflon (3 mm) & 400 μm and (d) Polyacetal (3 mm) & 600 μm .

We interpret these figures of radial segregation as resulting from a segregation process happening within the flowing layer, locating the large beads either at the top of the flowing layer (periphery of the drum), at the bottom (center of the drum), or at an intermediate level within the flowing layer (half-circle with $0 < r < R$). Figures 4 to 15 represent the radius of the segregation ring r/R versus the bead diameter ratio d_2/d_1 for growing tracer densities. The fit line is a guide for the eyes. Each mean radius r/R is associated to an error bar which is the standard deviation of the distribution of r_i . This standard deviation is a way to quantify the intensity of segregation: when the error bar is small, all the tracer particles are precisely located on a ring, if the error bar is large, particles are rather located all over the static bed.

1. Intensity of the segregation

Roughly, for each experiment, excepting the cases where $d_2/d_1 \leq 2$, the standard deviations of the distribution of r_i (represented as error bars on figures 4-15) are small, indicating that there is a well-defined segregation pattern. It is what we call a strong segregation process. In that case, r , the average of the distribution of r_i , is equal (or is very close) to the location where one large bead would preferentially be. In other words, the average value, r is equal to the value of the maximum of the r_i distribution [Fig. 16].

For the smallest size ratios, and especially for $d_2/d_1=1$, the standard deviations are large, and the segregation pattern is not really defined: the beads are spread more or less over the whole drum [Fig. 17]. We qualify the segregation process as weak. Consequently, if the distribution is non symmetrical, r/R can be significantly different from the maximum probability value [Fig. 16].

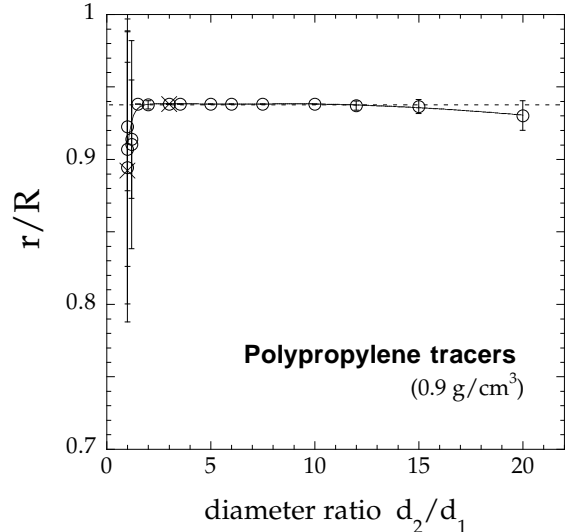


FIG. 4: Mean locations of 3-mm Polypropylene beads in a bed of small Glass beads: (O) 50 beads (corresponding to 3% volume fraction), (X) 17 beads (1%). Standard deviations of the distributions of the locations are represented as errors bars. All beads touching the periphery corresponds to $r/R=0.938$.

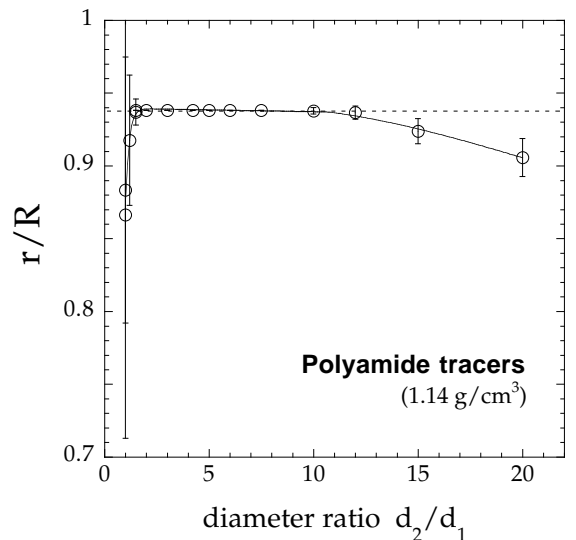


FIG. 5: Mean locations of 3-mm Polyamide beads in a bed of small Glass beads: (O) 50 beads (3%). Standard deviations of the distributions of the locations are represented as errors bars. All beads touching the periphery corresponds to $r/R=0.938$.

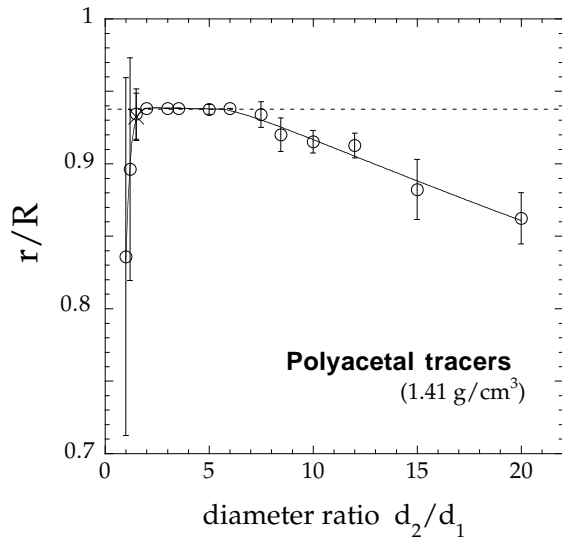


FIG. 6: Mean locations of 3-mm Polyacetal beads in a bed of small Glass beads: (○) 50 beads (3%), (×) 17 beads (1%). Standard deviations of the distributions of the locations are represented as errors bars. All beads touching the periphery corresponds to $r/R=0.938$. Experiment with size ratio 3.5 has been done at a larger speed 0.15 s^{-1} .

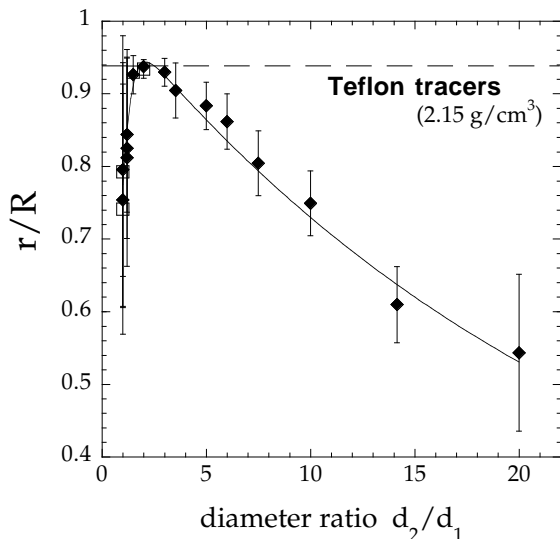


FIG. 7: Mean locations of 3-mm Teflon beads in a bed of small Glass beads: (◆) 50 beads (3%), (□) 17 beads (1%). Standard deviations of the distributions of the locations are represented as errors bars. All beads touching the periphery corresponds to $r/R=0.938$.

Here, we will discuss what information could be deduced on the segregation effects, coming from observations made on the focus of the segregation pattern. Tracers have been driven to their final location due to some forces exerted on them. We can separate them into a force due to the geometrical effect (difference of size), in forces due to the mass or density difference (buoyancy, weight), due to a shape difference, *etc.* The sum

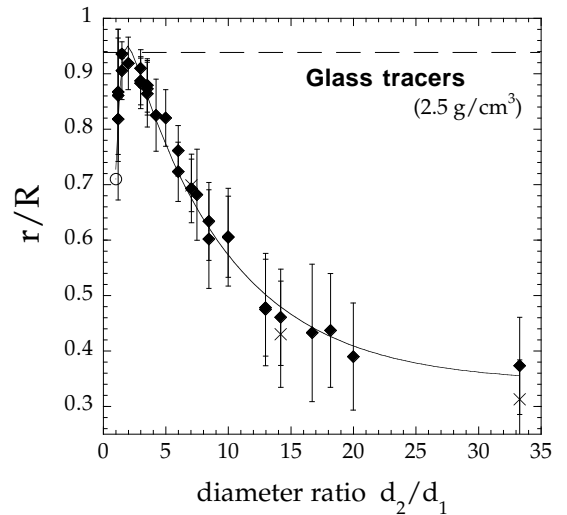


FIG. 8: Mean locations of 3-mm Glass beads in a bed of small Glass beads: (◆) 50 beads (3%), (×) 17 beads (1%), (○) theoretical homogeneous point ($d_2/d_1=1$; $r/R=0.71$). Compared to the figure presented in [6], some results have been added, and some have been slightly corrected by removing the tracers touching the caps when it was possible. Standard deviations (errors bars) are larger than for other materials due to the less precise hand of the experimenter at that time. All beads touching the periphery corresponds to $r/R=0.938$. The experiments with size ratios 7 (3%) and the upper result for size ratio 6 have been done at larger speeds, resp. 0.11 and 0.13 s^{-1} .

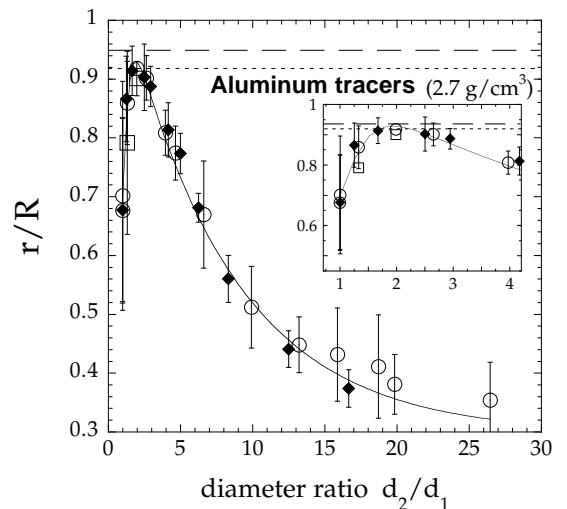


FIG. 9: Mean locations of Aluminum beads in a bed of small Glass beads. 3.97-mm beads: (○) 23 beads (3%) and (□) 15 beads (2%); 2.5-mm beads: (◆) 20 beads (0.65%). Standard deviations of the distributions of the locations r_i are represented as errors bars. All beads touching the periphery corresponds to $r/R=0.918$ for large beads, and to 0.948 for small beads. The curve is fitted using both sets of data. Inset is a close-up of the small diameter ratios.

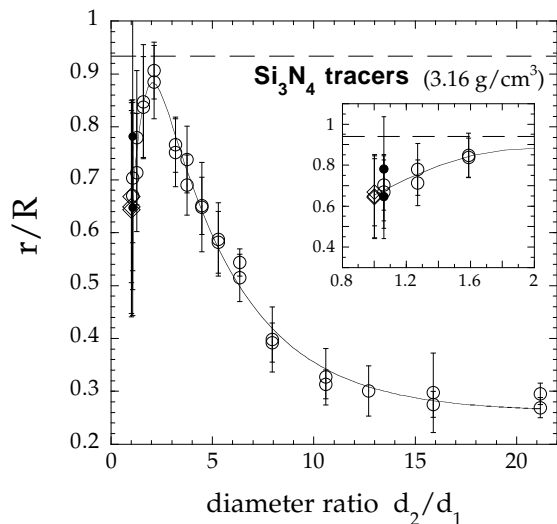


FIG. 10: Mean locations of Si_3N_4 beads in a bed of small Glass beads: (○) 10 beads (0.7%) of 3.175 mm, (◇) 10 beads (0.6%) of 3 mm (only for size ratio 1), (●) avalanches regime for size ratio 1.06. Standard deviations of the distributions of the locations are represented as errors bars. All beads touching the periphery corresponds to $r/R=0.934$. Each experiment has been done twice, except for size ratios 12.7 (once) and 1 (3 times).

of all these segregation forces determines the final location. Two cases have to be distinguished to interpret the focus of the segregation pattern. (1) When beads are at the top or at the bottom, the total force resulting of all segregation effects is not equal to zero. It is in balance with an iceberg effect near the free surface (tracers are emerging from the flowing bed), or with the effect of the static bed at the bottom of the flow (floor effect). The focus of the pattern depends on the intensity of the total segregation force. (2) When beads are at an intermediate level, the segregation effects balance at that level, and the total segregation force is equal to zero at that level. In that case, the dependency of the total force with the location z determines the focus of the pattern. When the total segregation force strongly depends on the location, the pattern is well-focused around the balance level.

In both cases, the intensity of the total force, or its derivative with the location z , has to be compared with the fluctuations of the tracer trajectories. We expect that the amplitude of the fluctuations depends on the size ratio d_2/d_1 , and on the thickness of the flowing layer in number of small beads (related to d_1/R). Moreover, fluctuations are enhanced when tracers interact.

Roughly, results show three tendencies: focus is very strong for tracers at the periphery, strong for high size ratios and weak for small size ratios. Very strong focus when tracers saturate at the periphery indicates that a non zero total segregation force is an efficient effect of segregation. This is not observed when tracers saturate at the center because it is masked by the interaction be-

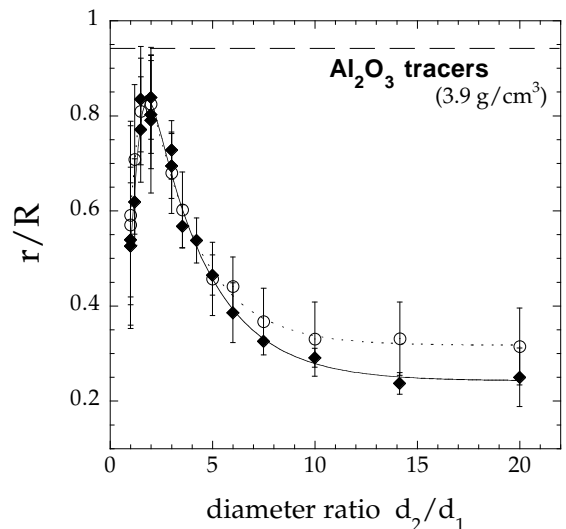


FIG. 11: Mean locations of 3-mm Al_2O_3 beads in a bed of small Glass beads: (○) 50 beads (3%), (◇) 10 beads (0.6%). Standard deviations of the distributions of the locations are represented as errors bars. The use of a larger amount of tracers slightly shifts r/R to larger values, but only at large diameter ratios. It also increases the standard deviations. The curve for 0.6% is the fit used in fig. 18.

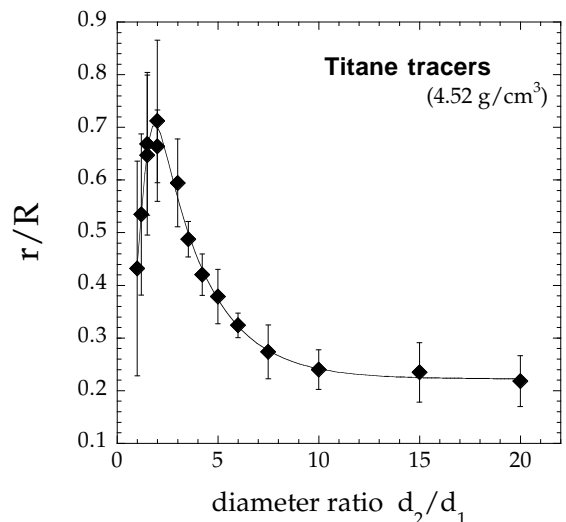


FIG. 12: Mean locations of 3-mm Titane beads in a bed of small Glass beads: (◇) 10 beads (0.6%). Standard deviations of the distributions of the locations are represented as errors bars.

tween tracers which creates fluctuations. This interaction effect is clearly seen in the case of Al_2O_3 tracers [Fig. 11] where two different fractions of tracers have been used (3% and 0.6%).

Several causes can explain the appearance of a weak segregation at small diameter ratios. It could be due to a decrease of the segregation force (or its z -derivative) or due to the enhancement of the fluctuations of trajecto-

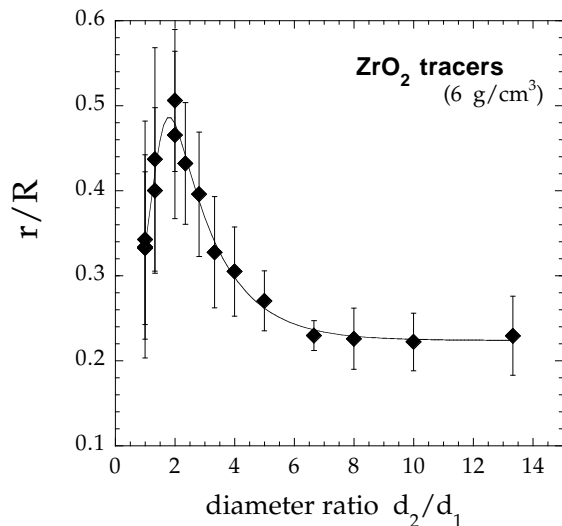


FIG. 13: Mean locations of 2-mm ZrO_2 beads in a bed of small Glass beads: (\blacklozenge) 25 beads (0.45%). Standard deviations of the distributions of the locations are represented as errors bars. Experiments with 1.3 and 2 size ratios have been done at a larger speed (0.15 s^{-1}).

ries. Fluctuations enhancement could be characteristic of small size ratios or could come from the fact that excessively large beads may have been used in the device. In fact, in our experiments, small diameter ratios are obtained by increasing the size of the small Glass beads of the bed, which leads to beds composed of large Glass beads (from 4 mm to 1 mm) compared to the drum diameter (48.5 mm). The flowing layer is then only 12 to 48 Glass beads long, which may not be enough to ensure a segregation process fully developed. Moreover, when the fraction of tracers is low, the thickness of the flowing layer is imposed by the size of the small Glass beads [27]. For such a geometry and according to [30], the thickness of the layer is around 6 beads for 1-mm Glass beads, to approximately 1-2 beads for 3-mm Glass beads. Such layers are close to the point at which the layer could not be still considered as a granular continuous flowing material, but more as a rolling layer of beads. In that case, trajectories of tracers and beads of the bed should be very chaotic and fluctuations are probably as large as the thickness of the layer.

To differentiate between the two possibilities explaining the enhanced fluctuations, we compare the 3 sets of experiments with different Steel tracers [Fig. 14], and the 2 sets with different Aluminum tracers [Fig. 9]: we found that for the smallest sizes of the tracers, the increase of the standard deviations at small size ratios still exists, although they imply the use of smaller sizes as the majority Glass beads and, consequently, thicker/longer flowing layers measured in number of Glass bead diameters. Moreover, in other devices (see section III), mixtures with a small diameter ratio look sometimes homogeneous (large standard deviations), whatever is the size of

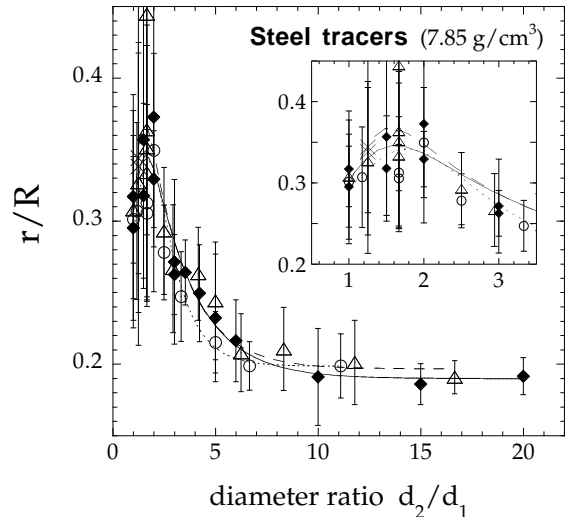


FIG. 14: Mean locations of Steel beads in a bed of small Glass beads: (\blacklozenge) 10 beads of 3 mm (0.6%), (\triangle) 15 beads of 2.5 mm (0.47%), (\circ) 72 beads of 1 mm (0.16%). Standard deviations of the distributions of the locations r_i are represented as errors bars. Solid fit for 3-mm bead (used in Fig. 18), dashed fit for 2.5-mm beads, dotted line for 1-mm beads. For 2.5-mm beads, experiments with size ratios 8.3 and 11.8 have been done at a larger speed (0.15 s^{-1}) and (\times) corresponds to avalanches regime. Decreasing the tracer size by a factor 3 does not change significantly the r/R curve and the standard deviation values.

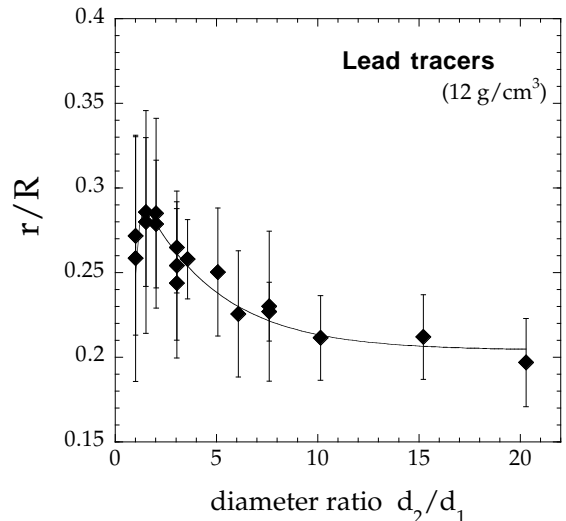


FIG. 15: Mean locations of 3-mm Lead beads in a bed of small Glass beads: (\blacklozenge) 10 beads (0.6%). Standard deviations of the distributions of the locations r_i are represented as errors bars.

the majority component particle compared to the thickness of flow. These two points prove that the weak segregation is not due to the use of excessively large beads as the majority component compared to the size of the drum. In conclusion, the segregation pattern is really focused by large values of the size ratio, and is not well-

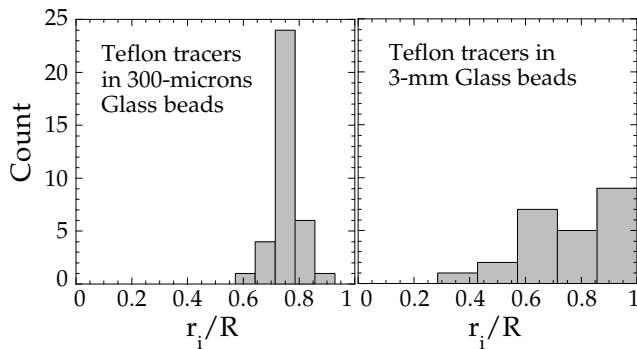


FIG. 16: Typical histogram of locations r_i of 3-mm Teflon beads in a bed of small Glass beads: On the left, the segregation is well-defined and the average corresponds to the most probable location, which is not the case for a diameter ratio equal to 1 (right histogram). Both r/R have similar values.

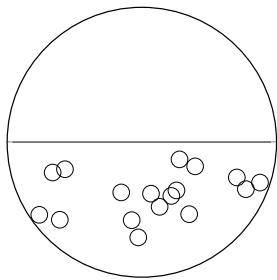


FIG. 17: Example of the locations of 3-mm large Si_3N_4 beads, in 3-mm Glass beads ($d_2/d_1=1$). The segregation does not really exist in that case.

defined for small size ratios (because of the variations of either the geometrical segregation force or the trajectory fluctuations). This point is independent of the value of the density ratio.

Consequently, density and size ratios do not play an equivalent role in determining the focus of the segregation pattern, even if they both equally participate in the choice of the mean location of the segregated tracers. Considering tracers *at intermediate location*, we deduce that the force of segregation due to a density ratio weakly depends on the location z (large standard deviations for $d_2/d_1=1$). But, data on standard deviations do not allow us to deduce informations about the gradient of geometrical force. In fact, small values of the standard deviations can be due to small fluctuations or to a strong dependency with the z location of the force coming from the geometrical effect of segregation (other arguments coming from r/R data (see end of the section II B 3) show that a z -gradient of geometrical force exists in the flowing layer of the drum).

We also would like to point out a possible statistical effect due to the spread patterns, which could influence the interpretation of the values of the mean location in terms of balance level. In fact, for the weak segregation,

the values of the mean locations r/R (for $d_2/d_1=1, 1.2$ or 1.5) are consistent with the other data obtained for larger size ratios [Figs. 4-15]. r/R also varies regularly with the density ratio for these small size ratios [Fig. 19]. Nevertheless, taking into account the asymmetry of the wide r_i distributions [Fig. 16], r/R could be significantly different from the maximal probability of r_i . If we assume that the maximum of the distribution is at the top (for light tracers) or at the bottom (for dense tracers), the mean value r/R is more or less shifted from these extreme values depending on the spreading of the distribution. The variation of r/R with the density ratio could then be a consequence of narrower or wider histograms, and not related to a variation of the maximal probability. The closer to 1 the density ratio is, the wider the distribution could be. The available data set is not statistically large enough to answer to this question, and further experiments have been planned. Consequently, the observed evolutions of the mean location r/R for small size ratios are compatible with two constant balance levels (at the top for light beads or at the bottom for dense beads).

In conclusion, we keep in mind that some initial values of the r/R vs d_2/d_1 curves may have to be shifted to 1 (for lighter materials than the Glass) and to 0 (for denser materials) to obtain the curves of the preferential locations (i.e. balance levels). We also know this is not the case when standard deviations are small (strong segregation) which is the case of most experiments. For most of the data, the evolutions of r/R are closely related to those of the preferential location.

2. Location of the tracers for each material

In these experiments, we vary the size of the small beads, which has been shown to be equivalent to varying the size ratio between the beads [6], and we vary the density of the tracers. Because of the beads available, we could not check if the position of the segregated circle depends on the density ratio or on the density. Nevertheless, we will assume in this paper that density and size ratios are the parameters controlling the location of the segregated large beads.

For each material, r/R presents the same evolution with d_2/d_1 : (1) A rapid increase of the mean location with the diameter ratio. (2) A long decrease of r/R with the diameter ratio [Figs. 4-15]. The increasing part of the curve is restricted to small size ratios (roughly <2) giving a maximum for r/R corresponding to size ratios between 1.5 and 2. We will see in the section III that indications of this complex variation of r/R versus d_2/d_1 , are found in other experimental devices.

(1)- The increasing part of the curve corresponds to the classical size segregation of large beads intensively studied by the granular community. The effect is stronger when size ratio increases. When passing from size ratio

1 to size ratio 2, the segregation radius increases, and eventually reaches the saturation value for low densities, i.e. all tracers touching the drum periphery. For larger densities, the maximum radius also occurs for size ratio close to 2, but the segregation ring is buried inside the drum.

(2)- The long decrease corresponds to tracers getting larger, and consequently heavier. They progressively sink into the bed by a push-away process. This new phenomenon has been named the intermediate and reverse size segregation by one of the authors [6]. It happens even for tracers lighter than the Glass beads of the bed. For the dense materials, the radius tends to saturate to its minimum value related to the thickness of the flowing layer.

These evolutions are similar to the evolution of the location of a large Glass tracer in a bed of small beads, corresponding to $\rho_2/\rho_1=1$ ([6] and Fig. 8). The theoretical point for $d_2/d_1=1$ (homogeneous media) would correspond to $r/R=2/3$ in a half-filled drum, but because we remove all tracers which are closer to the free surface than $0.18R$, a simple numerical calculation gives that the homogeneous media corresponds to $r/R=0.71$ in our data system. Compared to those of Glass tracers, the r/R curves are continuously shifted to the top for lighter materials, or to the bottom for denser materials [Fig. 18]: both evolutions of the location with the parameters ($d_2/d_1, \rho_2/\rho_1$) are continuous. Note that the data for Lead tracers are sometimes slightly above the data for Steel tracers. This may result of a little sinking of Lead tracers in the solid phase of the bed due to the very large density ratio [17]. These two continuous evolutions could appear surprising because we could have expected all beads denser than the Glass to be at the bottom, and all beads lighter than the Glass to be at the top. The balance at intermediate level for denser/lighter tracers than the Glass can not be simply explained by some variations of the compacity of the flowing bed. For example, they could correspond to relative densities between 1.85 (high packing fraction 0.74) and 1.25 (low packing fraction 0.5), but the range of densities of the tracers is far wider than that (0.9 to 12, or only from 2.15 to 6, if we limit this calculation to beads not touching the bottom or the surface). It means that density and size ratios both take a role in the selection of the mean location of the segregated tracers, as some authors already found [31].

From the r/R variations, we want to deduce the evolutions of the level where all segregation forces balance (preferential level). Both evolutions are linked when standard deviations are small (size ratios >2) but could be different for large standard deviations of asymmetric distributions (size ratios <2).

- For materials lighter than the Glass ($\rho_2 < 2.5$), we assume that the most probable value of the distribution of r_i could be the periphery. In that case, the preferential

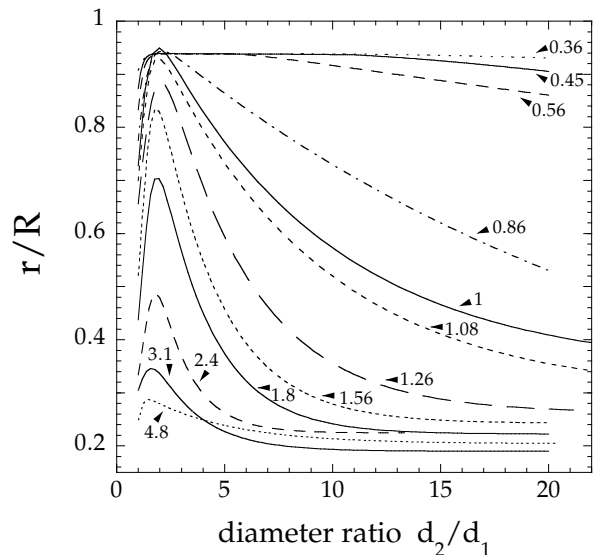


FIG. 18: All fits of the mean locations r/R of large beads of different densities (indexed 2) plunged in a bed of small Glass beads (indexed 1). Density ratios ρ_2/ρ_1 are written on the curves. Both density and size play equivalent role to determine the final location r/R . The denser the bead, the deeper its location.

location would first be constant up to size ratio 2, then decreases as d_2/d_1 increases in the same way as r/R . The complex evolution of r/R (increase then decrease) could be consequence of an initial asymmetric spreading of the distribution of r_i and not a real variation of the preferential location of the tracer.

- For denser materials ($\rho_2 > 2.5$), the expected most probable value will be close to the center of the drum, enhancing this increasing part of the curve. Consequently, an asymmetric statistical effect is not the reason for the increasing part of the r/R curve. The evolution of the balance location of the tracers really follows a complex curve (increase then decrease) similar to those of r/R .

One of the consequences of this complex variation is that two mixtures composed of the same materials, but with two different values of d_2/d_1 can show the same average pattern of segregation. In other words, two types of beads (for example Si_3N_4 beads with respectively 1.4 and 3 size ratios [Fig. 10]) will be found at the same place in a bed of Glass beads, leading to some possibility of mixing between two minority species inside a third majority component (here, the small Glass beads).

3. Influence of the density of the tracer on their location

Considering a given size ratio, the denser the large bead is, the deeper is its location [Figs. 4-15]. It is then possible to vary the vertical position keeping the geome-

try constant (same d_2/d_1) and changing the density of the tracer. The location of the tracers continuously evolves with the density ratio [Figs. 19, 20]. Denser (or lighter) beads than the Glass are not pushed out of the flow to the bottom (or surface), but do reach an intermediate level determined by the value of their density. The location of the segregated tracers is then driven by both density and size differences.

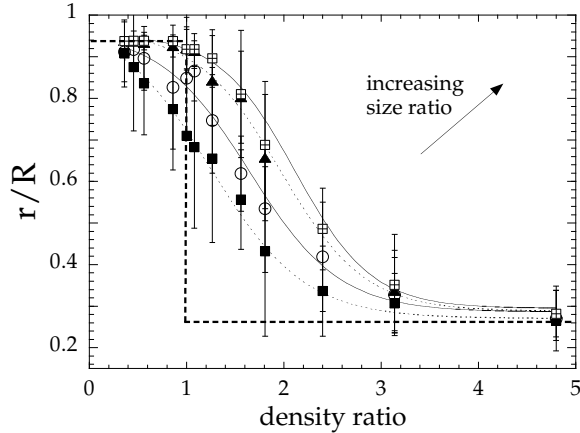


FIG. 19: Mean locations of tracers depending on the value of the size ratio: (■) 1 (more precisely 0.992-1.015), (○) 1.2 (and few points between 1.25-1.33), (▲) 1.5 (1.52-1.67), (◻) 2 (1.98-2.11). The lines are guides for eyes. The dashed step line is one theoretical possible curve for the preferential location for a size ratio equal to 1 (see section IIB 1).

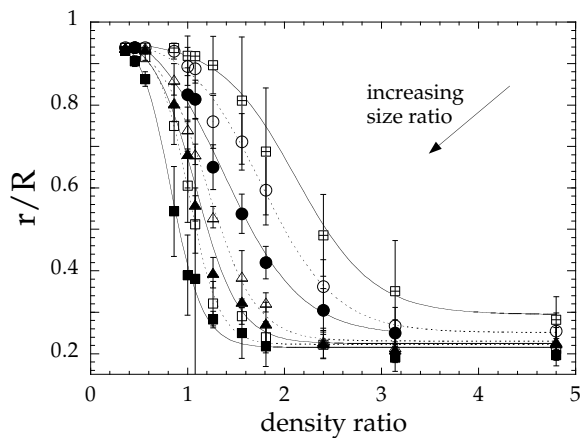


FIG. 20: Mean locations of tracers depending on the value of the size ratio: (◻) 2 (1.98-2.11), (○) 3 (2.94-3.175), (●) 4.23 (4-4.47), (△) 6 (6-6.67), (▲) 7.5 (7.5-8.33), (◻) 10 (9.92-10.58), (■) 20 (19.85-21.17). Data are averaged when several experiments have been done at the same density and size ratios. The lines are guides for eyes.

When increasing the density ratio, a large bead passes from the surface to the bottom. Roughly, this sinking appears earlier for the large size ratios than for the small ones because the tracers are already heavy when d_2 is large [Fig. 20].

But for the cases of size ratios 1, 1.2, 1.5, and 2 [Fig. 19], the sinking is delayed when the size ratio is bigger. It corresponds to the sharply increasing part of the r/R vs d_2/d_1 curves [Figs. 4-15]. For $d_2/d_1=1$, the evolution of r/R is continuous, and suggests that a pure difference of density could lead to a balance at an intermediate level inside the flow. This evolution of r/R is compatible with the results obtained by 2D simulations [17]. Quantitatively, our curve decreases more rapidly and goes further down than this numerical study, probably because the ability for the tracer to go down is reduced in the case of a 2D drum, compared to the case of a 3D drum. Because the preferential locations may be shifted from these r/R data by statistical effects (Sec. IIB 1), we also draw on the same graph a possible theoretical curve (dashed step line on Fig. 19) for the preferential location (periphery for lighter beads, center for denser beads than the Glass); the real curve of the more probable location is likely to be between this dashed step line and the r/R curve. Our actual set of data do not provide a means to obtain this preferential location curve and further investigations have been planned, before concluding if the balance at an intermediate level due to a pure difference of density is experimentally observed or not.

The density difference of materials is not the only reason for a large bead to be buried in the flowing layer. It could not explain the fact that large Glass beads or large beads made of lighter materials (Teflon, for example) can be buried. The observations prove that there is a mass effect involved in the segregation process as previously assumed [6]. The large tracers are heavy enough to push away their small neighbors and to go down. Thinking in terms of Fluid Mechanics, the mass effect can be interpreted by the fact that a Glass tracer bead is denser than a bed of small Glass beads containing at least 27% of void. This implies that, in the case where their volumes are really different, it is no longer possible to compare only one large and one small bead to determine what happens during the segregation. The density of the tracer has then to be compared to those of one small bead (for $d_2/d_1 \approx 1$) or to the density of the whole bed of small beads (for $d_2/d_1 \gg 1$). This implies also that tracers lighter than the density of the bed are always partially visible in surface. Their slight sinking is due to the weight of the non embedded part of the tracer (iceberg effect).

By varying the density ratio between the beads, only the mass effect changes. But, by varying the size ratio, both effects (geometrical and mass) change and it is more difficult to determine what part comes from each. When increasing the size, the mass effect tend to be dominant explaining the fact that the larger tracers get buried deeper. In that point of view, each tracer is affected by the mass effect, and by the dynamical sieving (geometrical effect) and its location is determined by the addition of these 2 effects. We choose to represent each effect by a force exerted on the tracer; the mass effect being the

sum of the weight and the buoyancy, the geometrical effect being the result of the actions of the small beads on the tracer (except the buoyancy). In that point of view, the mass effect depends on density and size ratios, the geometrical effect only on the size ratio. The balance is obtained where the two forces are equal, with opposite signs.

Considering a tracer flowing at an intermediate level within the flow, the mass and the geometrical forces exerted on it exactly counterbalance. This bead shows no vertical relative motion. If the balance is obtained at that level and not at another, it means that, at least, one of the two effects depends on the vertical coordinate z within the flow. Because the focus of the pattern is not strong for $d_2/d_1=1$, it is not very likely that the mass effect strongly depends on the vertical location. In fact, the buoyancy varies due to the variation of compacity inside the flow. Only a few studies present data on vertical gradient of compacity in the flowing layer of a drum [32, 33, 34, 35], but they concern 2D or quasi-2D experimental devices. Excepting the first layer of beads in which the compacity drops to 0, the variations in 2D are low (from 0.4 to 0.8) and increase with the rotational speed. From numerical simulations, an estimate of the maximal compacity variation in a 3D drum flowing layer is from 0.4 to 0.6 [36]. This compacity variation gives a variation of the buoyancy corresponding to a density difference of $(0.6-0.4)\times 2.5 \text{ g/cm}^3$ in small Glass beads. Although there is a large uncertainty on the compacity, these variations are far too small to balance variations of relative density of the tracers at intermediate level (from 2.15 to 6) (Sec. II B 2). Note that compacity variations have a small impact on the buoyancy force because they are proportional. On the other hand, it is possible that strong variations of the geometrical effect with z are related to these low variations of the compacity.

Moreover, we found tracers of the same size and of different materials located at intermediate levels (i.e. mass and geometrical effects in balance), although they induce very different values of the mass effect. This implies that they are submitted to very different values of the geometrical effect. Because they have the same size, these different values are due to the fact they are located at different levels. Consequently, the geometrical effect varies strongly with the location z .

From these two arguments, we conclude that the geometrical effect of the segregation strongly depends on the vertical location z within the flow.

The data also show that the density differences prevail for large and small density ratios [Figs. 19, 20]. Very light tracers are always at the surface whatever the value of their diameter (approximately for $\rho_2/\rho_1 < 0.5$) and very dense tracers are always at the bottom (approximately for $\rho_2/\rho_1 > 4$). For those extreme densities ratios, the value of its diameter does not influence the preferential location of one large bead. In these cases, there is no balance between the two effects: the mass effect always

dominates for very dense beads, or is acting upwards for very light beads, as the geometrical effect does. For all intermediate density ratios, the diameter ratio has an influence: the whole pair $(d_2/d_1, \rho_2/\rho_1)$ determines the location of the bead.

4. A few more remarks

Some complementary data are shown on curves r/R versus diameter ratio.

Two sets of experiments have been done with Silicone Nitride tracers [Fig. 10]. Both mean values r/R , each calculated on the ten r_i data, are similar, which is not surprising because of the small standard deviations of each set. The two sets show the good reproducibility of the results from one experiment to another. The average between both r/R is taken to fit the data.

Experiments with Steel beads involve 3 different diameters (1, 2.5 and 3 mm). They have been done to investigate the influence of the size of the tracer when it gets close to the center of the drum. No significant effects have been observed. The experiments using 1-mm beads give slightly smaller values of r/R , probably because 1-mm beads rolling on the bottom of the flowing layer have their center lower than the center of 3-mm beads.

Experiments with Aluminum beads have been done with 2 diameters (2.5 and 4 mm), showing no major difference either, except maybe when tracers are close to the center of the drum.

Both sets show that the size ratio is a relevant parameter when tracers are at intermediate level. Effects due to the finite size of the tracers appear only when tracers are very near the center or at the periphery.

Some experiments have been done with a different number of tracers to evaluate the influence of the volume fraction of the tracers on their final location. The influence of the volume fraction on the results has been previously tested on Glass tracers [6], and shows that the interaction between large tracers shifts the segregated locations toward the surface. This shift is combined with a spreading of the individual location around their average. Choosing d_2/d_1 such that a low fraction of large beads segregate at a deep intermediate level, a nearly homogeneous mixture has been experimentally obtained by increasing the fraction of large beads.

Consequently, two behaviors are expected when using a mixture of beads having different sizes and densities: (1) for dense large tracers the mixture tends to be more homogeneous when increasing their fraction; (2) for light large tracers, the mixture still segregates with large particles on the surface when increasing their fraction. These 2 evolutions are compatible with the results of numerical simulations [18] obtained on mixtures with 50% of each component, showing segregation for light large tracers and homogeneous mixing for dense large tracers. The

shift of the tracers towards the surface when increasing their fraction also explains that 50% of tracers with a size ratio 3, and a density ratio 1.5 will segregate at the surface [20]. In the same way, the value of r/R at low fractions can be used to estimate the segregation state of a mixture at a higher fraction. For example, mixtures with size ratio 1.14 (resp. 1.42), and density ratio 0.82 (resp. 1.53) which would lead to r/R values of 0.83 (resp. 0.75) in our drum device, give effectively homogeneous results at 50% [19]. For mixture with size ratio 1.14 (resp. 1.18) and density ratio 1.53 (resp. 0.69) which would lead to r/R equal to 0.6 (resp. 0.9), this gives down (resp. up) segregation of the large particles at 50% [19].

Here, we keep the fraction low ($<3\%$) and test the influence on Al_2O_3 Ceramic (3% and 0.6%) and a few other experiments indicated in the figures. Moreover, only 10 (resp. 20 and 25) beads were available in the case of Silicone Nitride, Titane (resp. 2.5-mm Aluminum and Zircon) corresponding to 0.7%, 0.6% (resp. 0.6% and 0.45%) volume fraction. We then needed to quantify the influence of the fraction to be able to compare all the sets of data obtained with these different fractions.

The results show that there is almost no influence of the fraction in this small range of fractions. Nevertheless, when all the tracers tend to be close to the center, the value of r/R is slightly smaller when using less tracers. The observed shift is small in our experiments but is strong enough to make two ‘parallel’ curves with different fractions of tracers cross each other when getting to small values of r/R (cases of Si_3N_4 -0.7% and Al_2O_3 -3% [Figs. 10 and 11]). The shift probably happens because too many tracers can not be all regrouped in the center. Moreover, the interaction between tracers enhances the trajectories fluctuations. It also prevents them from reaching the deeper point of the flowing layer because they tend to roll on each other (one tracer being in the flowing layer, one tracer being inside the solid rotating part). For that reason, only a few beads have been used in the case of Lead and Steel: 10 beads for 3-mm Steel and Lead beads (0.6%), 15 beads for 2.5-mm Steel beads (0.47%). But it was possible to use 72 1-mm Steel beads (0.16%), which do not interact much due to their smaller size. For an obstruction reason too, we expected that r/R would be slightly higher for smaller fractions, when all beads are close to the periphery. But, in this latter case, the shift of r/R would appear for higher differences of fraction due to the larger volume available at the periphery than at the center, and it has not been observed here. In conclusion, for these small fractions ($<3\%$), variations in r/R with the fraction are small and we do not take them into account in this study, except when the beads are all grouped at the center.

The rotation speed also varies from one experiment to another and the variations are randomly distributed for experiments done by hand. We do not notice any influence in the range of speed we use. This is not surprising because, for that range of size of drum and beads, we

expect that the thickness of the flowing layer is almost independent of the rotation speed [30] (an increase of the thickness of the flowing layer would geometrically imply a slight increase of r/R , significant only for beads located in the lower part of the flowing layer). Both sets involving Silicone Nitride tracers have been done with speeds around 0.085 s^{-1} and 0.15 s^{-1} , showing no difference, and no systematic relative order between the two sets.

Some of the experiments with the largest ‘small’ Glass beads (2-3 mm), which have usually been done with high rotating speeds, have also been done at the same speed as the others (0.085 s^{-1}). The flow is in the avalanches regime for these large beads. The results are very close when averaged, but locations of tracers are more spread and 10 measurements may not be enough to accurately determine the mean location r/R [Figs. 10 and 14].

III. DIAGRAM OF SURFACE SEGREGATION

One of the consequences of the complex shape of the curves r/R versus d_2/d_1 , (Sec. II B 2) is the fact that two large beads of the same density and of different diameters, can be segregated at the same level in a flow of small ones. For example, 3 and 4.7-mm Al_2O_3 beads would be at the same place in a bed of 2-mm Glass beads [Fig. 11].

In order to determine if that feature only happens in the drum device, or is due to the use of flows being only a few beads thick, we choose to work with three additional experimental devices in which the flow thickness varies. We indirectly explore the possibility for tracer particles of two different sizes (and of the same density) to be found at the same place in the flow. The 3 types of experiments are: flow in a channel, flow at the free surface in a half-filled quasi 2D disc (no bulk observations), and the build-up of a pile [Fig. 21].

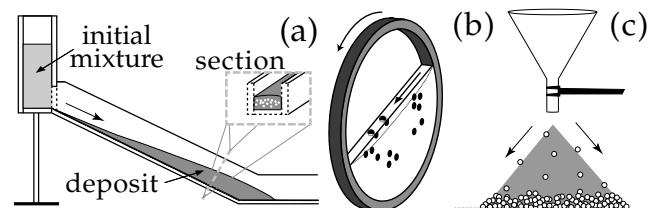


FIG. 21: Experimental devices for: (a) flows in a channel, (b) rotating a half-filled quasi 2D disc, (c) building a pile.

For practical reasons, the criterion we choose is the presence or absence of the tracers at the surface of the mixture. The tracers can be either visible at the surface, or buried inside the bed. ‘Visible’ corresponds to tracers being over the surface or partially embedded in the small particles. ‘Hidden’ corresponds to tracers inside the bed, whatever the location they have (intermediate or at the bottom). The aim is to look for the limit separating the two states ‘visible’ and ‘hidden’. When

on the limit, the mixture looks as if there was no segregation at all because the tracers do not appear to be preferentially at one location (neither inside, nor at the surface). On this limit, we do not differentiate between mixtures where there is really no segregation (homogeneous), and those where tracers are around a mean vertical position very close to the surface. This second case will be called ‘no apparent segregation’. It is sometimes possible to determine the difference between the 2 cases (by a cross-section inside the deposit), but most of the time, the experiments are not accurate enough to distinguish between a real homogeneous medium and a bad-focused segregation at a level close to the surface. From experiments done with Glass tracers in small Glass beads [6], no (at least apparent) segregation has been shown to occur in the channel for size ratio around 4.2, corresponding to a mean location of these tracers at approximately $r/R=0.8$ in the 3D drum device. A real homogeneous medium would correspond to $r/R=0.71$ (see section II B 2), which is not far from 0.8 taking into account the uncertainty of the visible-hidden criterion. By extension, we could assume that both really or apparently homogeneous cases are obtained for mixtures in which tracers would be around $r/R=0.7-0.8$ in the drum device (this hypothesis will be shown to be true in the section III B). The limit would then be close to the iso-position curves $r/R=0.7-0.8$.

In the following sections, we want to draw in the plane (ρ_2/ρ_1 ; d_2/d_1) the two fields (hidden, visible) of segregation. Then, we will examine if the shape of the limit between the fields is compatible with the fact that two different types of tracer of the same density can be found at the same level in a bed, i.e. if the shape of the limit is or is not a single-valued function from $[\rho_2/\rho_1]$ to $[d_2/d_1]$.

A. Experimental devices

In all these experiments, the initial homogenous mixture is composed of a small volume fraction of tracer particles indexed 2 (2% for disc and piles, 10% for flows in the channel) in a bed of particles indexed 1. Large and small particles are either beads (Glass, Steel, Sugar, Diakon), or angular particles (Sand, Silicone Carbide, fragments of Glass, of Sugar, of Wood Nut). Most of the particles used in the previous drum experiments are not adapted here, for size (in disc) or available quantity (in piles, channel) reasons. There is no systematic use of one type of particles to play the role of the tracers, or of the majority. Choices have been made to obtain various density and size ratios, preferentially with spherical particles when it was possible. Densities of the particles are reported in table II, and the diameters used on the graphs are the mean sizes of the given ranges, obtained by sieving.

TABLE II: Particle densities and sizes

Materials :	density (g/cm ³)	sizes or ranges of sizes (in μm if ‘mm’ not written)
Glass beads	2.5	45-90; 70-110; 150-212; 180-212; 150-180; 150-250; 300-400; 300-425; 425-600; 600-710; 600-800; 600-850; 710-800; & 90; 150; 180; 200; 212; 250; 300; 355; 400; 425; 500; 600; 710; 800; 850; & in mm: 1.5; 2; 3; 4; 5; 7.5 in mm: 0.85-1; 1-1.18; 1.4-2
Sugar balls	1.6	
Steel beads	7.78	4.7 mm
Diakon particles (ovoid)	1.14	106-212; 300-400; 300-425; 600-850; 625-850
Silicone Carbide fragments	3.2	150-212; 212-300; 300-425; 400-500; 500-600; 600-850; 850-1180; & 600
Wood Nut fragments	1.35	300-355; 450-560; 450-800; 630-710; 600-800; 710-800; 1-1.7 mm
Sugar fragments	1.6	150-250; 150-300; 300-425; 600-850; 850-1000; 1-2 mm
Sand	2.6	600-850
Glass fragments	2.5	300-425; 425-500

1. Rotation in a quasi 2D half-filled disc

For these experiments, we use a clear plastic disc whose internal dimensions are 3.5 mm in thickness, and 81 mm in diameter. The system is close to be 2D for largest tracers, but is 3D with walls effects for smaller particles. The disc is half-filled and rotates around its axis, placed horizontal. Rotation is obtained with a motor, two speeds have been tested: 0.033 s^{-1} which corresponds to successive avalanches and one speed between 0.8 and 0.16 s^{-1} chosen to obtain a continuous flow and a plane free surface. We observe through the walls what species is present at the surface of the flowing layer, and what species ends up at the periphery of the drum. One limitation of this device is the formation of plugs which modify the radial location of large particles when their size is close to the thickness of the drum [6]. Strong electrostatic effects are noticed, especially for some small particles, depending on the material they are made of (Diakon, Wood Nut). Consequently, we do not expect accurate results with this device and we focus on the general behavior. Considering the criterion “being visible on the surface during the flow”, we found that the two sets of results obtained with the different speeds are strictly identical: the segregation process in a continuous flow or in a single avalanche is the same. We thus only draw 1 point for each couple (ρ_2/ρ_1 , d_2/d_1) on the figure 22 which represents the type of segregation vs both density and size ratios. The three following figures will be discussed together in the section III B.

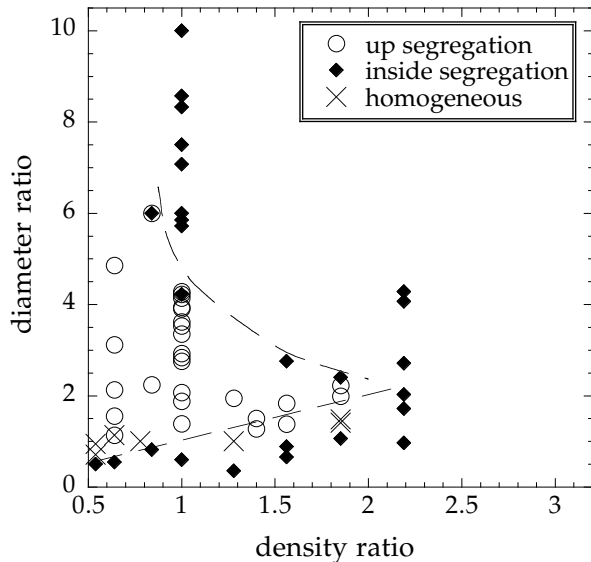


FIG. 22: Fields of surface segregation in the disc: (○) tracers flowing on the surface, (◆) totally embedded or at the bottom of the flow, (×) mainly embedded but still visible at the surface or homogeneous. Ratios are characteristics of the tracers on those of the majority component. Two symbols added means that the result is likely to be one of the two cases, or that two experiments have been done giving no clear determination. The limit between the two fields is the same in pile and channel experiments graphs.

2. Formation of a pile

When a mixture of two types of particles flows from a funnel, the heap formed beneath exhibits features of segregation [6, 37, 38]. In our experimental system, the mixture slips along the funnel walls, and no segregation happens in the funnel [39], contrary to flows where the mixture is sheared [40]. The segregation results from phenomena taking place in the flow along the pile slopes: dynamical sieving, rolling of the beads on a relatively smooth surface [41] and, burying of the heavy particles into the bed. The particles located at the surface of the flowing phase go further than the others and accumulate in a ring around the pile base. Consequently, an up-segregation of tracers during the flow corresponds to the formation of this basal ring of tracers, contrary to a segregation at an intermediate level for which tracers stop on the sides of the pile, and to a bottom segregation for which tracers stop near the summit of the pile (for pictures see [6]).

We place 250 to 850 g of a homogeneous mixture in the funnel by putting tracers between successive fine layers of the main particles. The funnel is placed at 12 cm above a rough plane, and the aperture is opened: the pile builds up. The flux ranges between 10 and 30 g/s depending on the type of particles used. The flow is then continuous along the slope of the pile in the first stage, then proceeds as successive axisymmetric avalanches when the

pile gets bigger. Limitations are the use of a large quantity of particles which are not always available in every size and density, and occurrences of blocking of the funnel for beads larger than 5 mm. Contrary to the channel experiments (Sec. III A 3), the flowing layer is always thin, and vanishes to zero along the slope when the pile gets bigger, allowing the possibility to see buried particles. The criterion chosen here corresponds to the basal ring formation or not, because large tracers are always visible when the flowing layer becomes very thin. We also take into account the fact that the tracers are grouped closer to the center or closer to the periphery. That gives the two fields of segregation (up and inside) in the figure 23, which will be discussed in section III B.

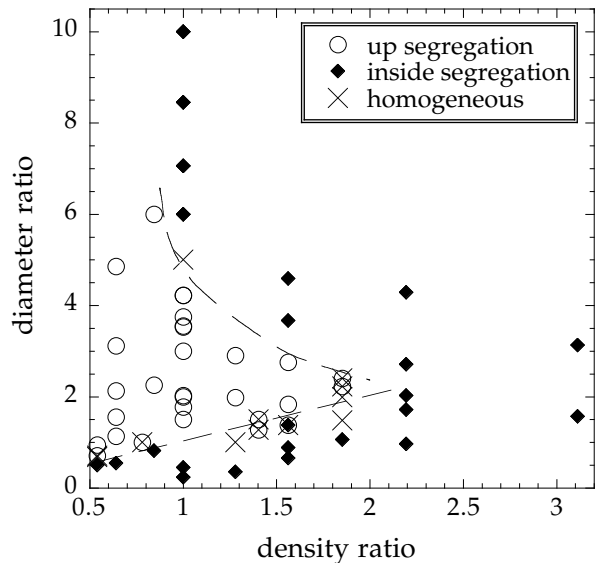


FIG. 23: Fields of surface segregation in piles: (○) tracers making a bottom ring on the pile, (◆) embedded in the center of the pile, (×) piles looking homogeneous. Two symbols added means that the result is likely to be one of the two cases, or that two experiments have been done giving no clear determination. Ratios are characteristics of the tracers on those of the majority component.

3. Flow along a slope

Chute flows have been experimentally and numerically studied [1, 6, 16, 42, 43] for mixtures of different size and density particles. We choose to study slow frictional flow, and avoid rapid flows. The chute is a 1 m long, 6 cm wide channel inclined with an angle of about 27° for beads (from 26.5° to 30°), and up to 33.5° for angular particles, chosen to produce a frictional flow [Fig. 21]. The bottom is made rough by gluing Glass particles or angular fragments on it. Glued particles are chosen to have the same size as the majority component of the mixture. The starting box at the top of the slope is filled with ~ 1 kg of a homogeneous mixture obtained

by placing alternating fine layers of both species. The main limitation is the large quantity of particles required. The opening of the box produces a thick flow with a mass flux of about 400 g/s. The flow stops when the channel becomes horizontal. Direct observations of the flow and of the internal structure of the deposit show that the deposit is the result of a strong deformation of the main flow when it stops. However, the surface of the flow stays on the surface of the deposit when it stops: in spite of the deformation, there is little doubt in diagnosing the location of a species above or under the surface of the flow by looking only at the deposit. We took as criterion whether the tracers are visible or not at the surface deposit, leading to the two fields of segregation presented in figure 24, which will be discussed in section III B.

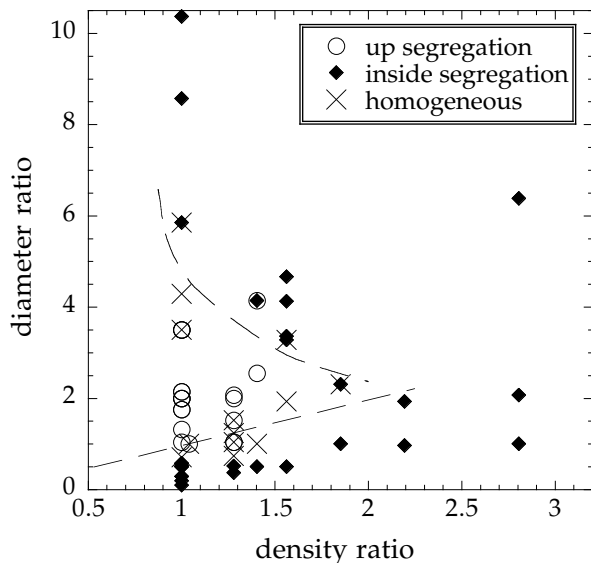


FIG. 24: Fields of surface segregation in the channel: (○) tracers cover the surface, (◆) are embedded in the deposit or at the bottom, (×) are both inside and at the surface (homogeneous). Two symbols added means that the result is likely to be one of the two cases, or that two experiments have been done giving no clear determination. Ratios are characteristics of the tracers on those of the majority component.

Moreover, in our experiments, particles have different shapes. Glass fragments are more angular than Silicone Carbide fragments, Wood Nut or Sugar fragments. Sand angularity is intermediate between beads, and Silicone Carbide fragments. In every case, the sizes in the 3 axes of the particles are similar (typically a factor 1-2 between smaller and larger sizes for Sand, a factor 2-3 for Silicone Carbide, Sugar and Wood fragments, a factor 5 for Glass fragments). When mixed, all these particles give several angularity contrasts. We expected an influence of the contrast on the final segregation pattern, and on the ratio values corresponding to the limit.

On the figure 25, we can directly observe the effect of a difference of shape on the segregation. Different particles

have been used, mixed either with Glass beads or with angular fragments of Glass. Up, inside, or homogeneous mixtures have been found depending on the size ratio used.

First, we find that when Glass particles with different shapes and of the same size (defined by the same sieving interval) flow together, angular particles segregate at the surface (Glass fragments and beads (○) on Fig. 25). These results seem in accordance with simulations done on the shaking of granular elongated particles [44] which show different possible segregation types depending on the elongation of the particles.

Second, we notice that when tracers are more angular than the particles of the bed (cases of SiC fragments or Glass fragments, mixed with Glass beads in Fig. 25), the homogeneous state (×) seems to be obtained for smaller size ratios than expected [21] (dashed line and (+) in Fig. 25). For Glass particles, the shift in size ratio due to the angularity (represented by an arrow) extends roughly from 1 to 0.7. We deduce that the Glass fragments behave as approximately 1.4 bigger beads. Vice-versa, when tracers are less angular than the particles of the bed (case of SiC fragments mixed with Glass fragments in Fig. 25), the homogeneous state seems to appear for greater tracers than expected. The directions of the shift are also compatible with the fact [see Fig. 24] that the 4 (×) under the bottom dashed line correspond to tracers more angular than particles of the bed, and that the 3 (×) above the bottom dashed line correspond to tracers less angular. Both deviations suggest that an excess of angularity pushes the more angular particles to segregate at the surface, although the gap between experiments (for density ratios different from 1) do not allow us to conclude if a shift really exists or not.

Nevertheless, using angular particles whose 3 dimensions are close (neither splinters, nor discs), this shift remains small in amplitude and explains why data obtained with various shapes of particles are regrouping in two distinct zones.

B. Recursive limit between the two fields of segregation

On each set of experimental results, two domains can be clearly defined in the plane (ρ_2/ρ_1 ; d_2/d_1) [Figs. 22-24]. The existence of these two distinct zones shows that the segregation pattern is a function of the size and density ratios and not of the values of individual diameters and densities. Moreover, the results obtained in the three systems are very similar: the same dashed curve is plotted on the three channel, pile and disc graphs, showing that the segregation process during the flow does not depend on the geometry inducing the frictional granular flow. The top part of the limit curve has been chosen as a compromise using all sets of data. The bottom part is the line $\rho_2/\rho_1=d_2/d_1$ corresponding to the homogeneous mixtures obtained by Alonso et al. [21], for very small

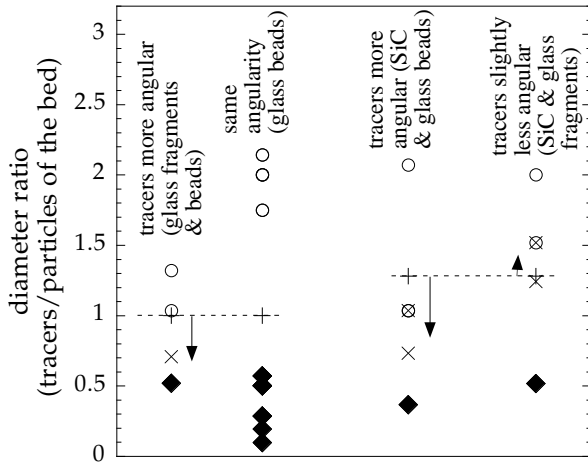


FIG. 25: Channel experiments for 4 mixtures having different angularity contrasts: (○) tracers cover the surface, (♦) tracers are embedded in the deposit or at the bottom, (×) homogeneous state, (+ and dashed line) theoretical homogeneous state for particles of the same shape. When angular and spherical particles of the same ‘size’ and density are mixed, there is an up-segregation of angular particles. The use of more angular particles as tracers (resp. as particles constituting the bed) seems to shift (indicated by arrows) down (resp. up) the diameter ratio for which the mixture remains homogeneous.

fractions of tracers. We can also notice that the processes happening during the shaking or during the shearing of a granular medium are different. The results for our three devices are quite identical, but they are rather different from those recently obtained in a vertically shaken box [8].

On each graph, the limit is chosen as one curve passing between the two types of segregation pattern. More precisely, this limit passes through the experiments with no (at least, apparent) observed segregation (marked as “homogeneous”). For all devices, the shape of the limit is a recursive curve and not a curve of a single-valued function from $[\rho_2/\rho_1]$ to $[d_2/d_1]$. The area where the tracers are visible in surface (○) is surrounded by the area where the tracers are buried (♦). Consequently, a vertical line (ρ_2/ρ_1 constant) can cross twice the recursive limit between the two fields of segregation, depending of the value of ρ_2/ρ_1 . For these density ratios, the tracers are under the surface of the bed when they are small or when they are very large, and they are on the surface when they are medium size. Consequently the mean position of the tracers evolves up then down, in an analogous way of the r/R curves [Figs. 4 to 15] when increasing the size ratio. This implies that one can find two tracers with different diameters and the same density, which segregate at the same vertical mean location. We deduce that the ‘bell’ shape of the previous r/R vs d_2/d_1 curves [Figs. 4 to 15], is not due to the use of excessively large beads constituting the bed of granular matter in the drum device (Sec. II B 2). This particular shape of the r/R location

curve is really a consequence of the variation of the diameter ratio.

Considering only the case where $d_2/d_1 > 1$ [Figs. 22-24], the double intersection of the limit is characteristic of dense tracers, as the ‘bell’ shape is characteristic of materials denser than the Glass [Figs. 4 to 15]. This double intersection exists only for density ratios less than about 1.8 (between 1.6 to 2.2). Tracers having larger density ratios are never visible in surface. From the previous drum data (Sec. II), we deduce that it corresponds to tracers which never go higher than $r/R=0.7$ (between 0.84-0.6) whatever their size are. That implies that visible locations correspond to approximately $r/R > 0.7$ (strictly $r/R > 0.84$) in the drum.

Vice versa, the ‘bell’ shape of all the r/R vs d_2/d_1 curves from the previous drum experiments, suggests that the iso vertical levels in the flow (i.e. same r/R value in the drum) have a recursive shape when represented in the plane (ρ_2/ρ_1 ; d_2/d_1). Using the results of the section II, we can draw the iso-positions of tracers curves in this plane for several choices of r/R [Fig. 26]. These curves correspond to the intersections of the r/R versus (ρ_2/ρ_1 , d_2/d_1) surface by planes $r/R=\text{constant}$: the iso- r/R curves have effectively recursive shapes. Moreover, the limit defined from disc-pile-channel experimental data, is close to iso curves $r/R=0.7$ and 0.8. This shows that the limit corresponds roughly to one isolation. As $r/R=0.71$ corresponds to a homogeneous medium in our data system (see section II B 2), it is not surprising that the limit corresponds to these particular values of r/R .

From the drum experiments, we know that the iso-position curves are not symmetrical in terms of intensity of segregation: the lower part (close to line $\rho_2/\rho_1=d_2/d_1$) corresponds to large standard deviations, and the upper part corresponds to small standard deviations. This fact is compatible with direct observations on channel experiments (from cross-sections) in which mixtures look rather homogeneous on the lower part of the curve, and rather segregated at an intermediate level close to the surface for the upper part. These observations are only visual, but seem to confirm that large size ratios enhance the focus of the segregation pattern, whereas both size and density ratio determine the average location of segregated tracers.

IV. CONCLUSION

In this study, we found that large tracers segregate either at the bottom, at the surface or, in most cases, at an intermediate level in the flow. This last location creates well-defined ring patterns of segregation in a half-filled rotating drum. Reversing between surface and bottom segregation is also observed in other devices (chute flow, pile building) where the granular matter is also subject to a frictional flow.

In the drum device, the radius of the ring varies con-

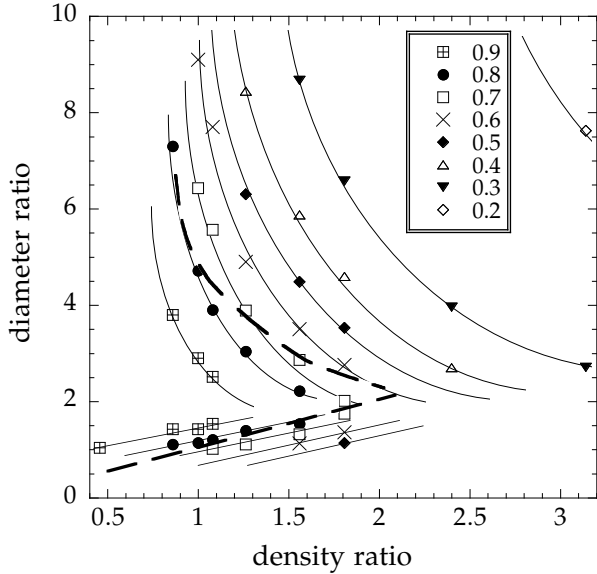


FIG. 26: Curves linking characteristics of tracers which segregate at the same r/R location in the drum experiments (values of r/R in the inset). Symbols are values taken from the fits of r/R vs d_2/d_1 , lines are just guides for eyes. Iso- r/R 0.7 and 0.8 are close to the limit (thick dashed curve) between fields of segregation observed in pile/disc/channel experiments.

tinuously with both the density and size ratios of tracers compared to the particles of the bed. It is then possible to consider that a difference of size has an equivalent effect on the tracer location than those of a difference of density (and vice-versa). Compared to size segregation of beads of the same density, the progressive sinking of large tracers at high size ratios is enhanced (resp. delayed) for denser (resp. lighter) tracers.

These facts are interpreted by the existence of two segregation processes affecting the tracers: a geometrical effect (dynamical sieving process) and a mass effect (push-away process). We could have expected that the density difference between the particles is responsible for one effect, and that the size difference is responsible for the other one. But, a size difference introduces the two effects of segregation (geometrical and mass), while a density difference implies only the mass effect. The push-away effect allows heavy beads to go towards the bottom of the flow. The geometrical effect drives large beads towards the free surface. When tracers segregate on a ring, these two effects exactly balance at a well-defined vertical level inside the flow. The level is the equilibrium location for

this tracer. The segregation of the tracers appears consequently to the regrouping of all these identical tracers at the same equilibrium location.

In the size segregation between beads of the same density, the geometrical effect is dominant at small size ratios (large beads segregate at the surface, as usually observed); the mass effect is dominant at very large size ratios (large beads segregate at the bottom); both geometrical and mass effects balance at intermediate ratios (large beads segregate at an intermediate level). The variation of the relative intensity of both effects explains the progressive reversing of the location of large beads when increasing the size ratio.

Because the balance happens at one particular level when a ring forms, we deduce that at least one of the two segregation effects depends on the vertical coordinate in the flow. The evolution of the ring pattern gives two types of information: (1) the focus of the pattern determines the intensity of the segregation process. It is observed to be strong for high size ratios, weak for small size ratios, whatever the density ratio is. (2) The variation of the radius of the ring shows that different mass effects can be in balance with geometrical effects due to the same size ratio, but acting at different levels in the flow. Data on the focus and the radius allow us to conclude that the geometrical effect depends strongly on the size ratio, as expected, but also on the vertical location in the flow. While the mass effect depends mostly of the size and density ratios.

Consequently, the two effects (mass and geometrical) implied in the *size segregation of beads of equal density* can be separated when using the whole set of data acquired on the locations of *segregated beads of different densities and sizes*: (1) Considering two tracers (of different sizes and densities) at the same location, the difference of geometrical effect due to the size difference is exactly balanced by the difference of mass effect. (2) In the same manner, considering two tracers of the same size, being at two different locations (which is possible if they have different densities), the difference of geometrical force due to the variation of the location is exactly balanced by the difference of mass effect. By modeling the mass effect, we will be able to calculate the geometrical effect function of both location and size ratio. This is currently under development in a model of the geometrical effect responsible of the segregation, using the whole set of experimental r/R equilibrium locations of the tracers.

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