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(54) **METHOD AND APPARATUS OF SEPARATING PARTICULATE MATERIAL AND PARTICULATE MATERIAL**

(57) The present invention relates to an apparatus and to method of separating particulate material with particle diameters in the range below 1 mm to around 1 μ m, the particulate material having an initial poly-dispersity of less than +/- 10%, the method including the steps of:
a) inserting the particulate material into a drum having an axis so that the drum is partially filled with the material,
b) rotating the drum with the particulate material therein about its axis, with the axis of the drum being generally horizontal, at a rate such that a free surface of the par-

ticulate material shows continuous flow dynamics,
c) continuing the rotation at least until radial micro-segregation of the particulate material is achieved, and
d) extracting particles from at least one zone of the micro-segregated particles having a poly-dispersity of substantially less than the initial poly-dispersity. The invention further relates to particulate material having a poly-dispersity of substantially less than the initial poly-dispersity.

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Description

[0001] The present invention relates to a method and to an apparatus of separating particulate material as well as to particulate material.

[0002] Particulate material, such as spherical glass beads, can be used as spacer elements, as lenses for optical fibers, as optical tweezers etc. For example, in the production of lenses for optical fibers lens material having a mono-dispersity as low as possible is sought. For this reason glass beads are separated into various size ranges for their different use as lenses.

[0003] The separation of the glass spheres into batches of material can take place using a variety of different methods. For example sieves having an accuracy of 5% are commercially available to separate the glass particulates. However, for certain applications a 5% disparity in size is not sufficient to be commercially viable.

[0004] For this reason it is an object of the present invention to provide a method and an apparatus of separating particulate material in which the disparity of sizes is reduced, and using which the particulate material with a very low poly-dispersity can be produced in a more cost effective manner.

This object is satisfied by a method according to claim 1.

[0005] According to the method in accordance with the invention the particulate material has particle diameters in the range below 1 mm to around 1 μm , and the particulate material has an initial poly-dispersity of less than +/- 10%, the method includes the steps of:

- a) inserting the particulate material into a drum having an axis so that the drum is partially filled with the material,
- b) rotating the drum with the particulate material therein about its axis, with the axis of the drum being generally horizontal, at a rate such that a free surface of the particulate material shows continuous flow dynamics,
- c) continuing the rotation at least until radial micro-segregation of the particulate material is achieved, and
- d) extracting particles from at least one zone of the micro-segregated particles having a poly-dispersity of substantially less than the initial poly-dispersity.

[0006] By means of the radial and axial micro-segregation in the rotating drum particles are separated into different zones within the drum. In these zones the particles have a poly-dispersity, which is substantially less than the initial poly-dispersity and advantageously is less than 2% in order to thereby achieve a separation of the particulate material into batches of particles having a very low poly-dispersity, otherwise unachievable in a cost-effective manner.

[0007] On generation of the plurality of zones of particles having a low poly-dispersity, the different zones can be extracted from the drum using a variety of different methods.

[0008] The particles of interest can be separated in a very cost effective and expedient manner which makes the method according to the invention commercially viable.

[0009] Preferably the particulate material comprises spherical particles. Using spherical material as the starting particulate material, the method can be employed in a particularly cost effective manner. Moreover, the achieved poly-dispersity of the micro-segregated particles can be significantly less than for non-spherical material using the same method and/or apparatus parameters.

[0010] In this connection it should be noted that the spherical shape of the particulate material is the preferred shape of the particle, the method does, however, principally work with particles that are rounded but not truly spherical, in which case the reference to the diameter will be understood to mean an average cross-sectional dimension in three mutually perpendicular directions.

[0011] It must further be noted that poly-dispersity refers to a measure of the heterogeneity of sizes of particles in a mixture of particulate material. In the present case this means that the polydispersity range comprises 90 % of the particle diameters.

[0012] Advantageously the particulate material comprises particles having the same density.

[0013] In this connection it must be noted that this will generally mean that the particles are of the same chemical composition, but also means that the method is applicable to particles of different materials, providing these have the same or substantially identical densities.

[0014] Preferably a fluid is additionally added to the partially filled drum. The fluid can enhance the continuous flow dynamics and thereby the formation of radial micro-segregation.

[0015] In this way the fluid can be added to the drum so that the drum is filled with fluid and particulate components. In this way the fluid at least fills the spaces (also known as interstitial spaces) present between the particulate material. The fluid may, for example, take up 10 to 30% of the filling volume of the drum.

[0016] In this connection it should be noted that when reference is made to a volume in this application then this relates

to the filling volume of the drum. The filling volume is the amount occupied by the particulate material and the interstitial fluid.

[0017] Advantageously the fluid is a gas or a liquid that is selected from the group comprising liquids capable of freezing, liquid phases of alcohols, liquids capable of easily vaporizing, liquid waxes, as well as liquids which permit the dissipation and screening of electro static charges.

[0018] This beneficially promotes the easy removal of the separated particulate material of interest. For example, liquids capable of freezing permit the distribution in the at least one zone of the micro-segregated particles to be frozen in place so that the particles in the at least one zone can readily be separated from particles in other zones, by removing each zone in the frozen state of the system.

[0019] The use of liquids which evaporate rapidly enhances the carrying out of the method and allows the liquid used to be readily separated from the particles by evaporation.

[0020] Liquids capable of dissipating and screening electrostatic charges prevent electrostatic charges from modifying the distribution of the particles in the at least one micro-segregated zone, such liquids can, for example, comprise water and diverse alcohols.

[0021] Additionally or alternatively liquid waxes can be used which are caused to solidify following the achieved state of radial micro-segregation in order to produce the desired zones from which particulate material can be extracted with the desired poly-dispersity.

[0022] Advantageously the drum is partially filled with particulate material when the particulate material takes up 10 to 90%, preferably for 20 to 70% and especially of 30 to 60% of the filling volume of the drum. Such filling volumes have found to produce the desired micro-segregation of the particulate material.

[0023] This filling volume has been found to be advantageous to bring about the desired continuous flow dynamics of the free surface of the particulate material.

[0024] This range of values is particularly beneficial for a drum having a diameter in the range of 2 to 4 cm and a length of approximately 50 cm.

[0025] Advantageously the speed of rotation of the drum is selected such that the surface shows continuous flow. This speed is above the threshold of the individual avalanche regime and below the threshold for the grain transport being dominated by saltation.

[0026] For example, for a drum having an inner diameter in the range of 2 to 4 cm and a length of approximately 50 cm, the speed of rotation conveniently lies between 12 and 35 rpm. For a drum having an inner diameter of 40 mm the speed of rotation corresponds to a range of 12 to 25 rpm. These speeds of rotation have been found advantageous in the promotion of radial micro-segregation.

[0027] Preferably particles having a larger diameter than the particulate material of interest are further provided in the drum.

[0028] The larger particles can optionally be added on each side of a region containing particulate material.

[0029] The advantage of the larger particles is that the presence of the larger particles may enhance the micro-segregation into the zones having smaller poly-dispersity.

[0030] The larger particles are filled into the drum so that they occupy 20 to 30% of the filling volume of the drum.

[0031] Advantageously a surface of the drum is provided with a surface roughness to enhance at least the radial micro-segregation.

[0032] This is an additional technique which may promote the micro-segregation of the particulate material.

[0033] The surface roughness of at least one of a jacket surface of the drum or an end surface (top and base surface) of the drum is advantageously selected in the range of 2 to 2000 μm rms. For particulate material with small particle diameters of 1 to 100 μm , a smaller surface roughness of e.g. 2 to 200 μm rms is selected, whereas for particulate material with particle diameters in the range of 100 to 1000 μm rms, a larger surface roughness of e.g. 200 to 2000 μm rms is selected.

[0034] Preferably the poly-dispersity of the particulate material in the at least one zone of the micro-segregated particles has a poly-dispersity of less than 2%.

[0035] So far as the present applicants are concerned, the present invention is the only way of economically obtaining a relatively high yield of particles having a narrow poly-dispersity of less than 2%.

[0036] Preferably particles having the desired poly-dispersity are continuously or discontinuously extracted from the at least one zone. Thereby particles can be removed either continuously from the at least one zone and batches of material are then prepared in a subsequent method step. Alternatively the particles can be removed batch wise from the at least one zone.

[0037] The removal can optionally take place during the rotation of the drum or when the drum is stationary. This method recognizes that a continuous process could be achieved if new material having a higher poly-dispersity is continuously or discontinuously added to the drum.

[0038] Preferably the continuous or discontinuous process comprises a scraping or a suction action. Such scraping or suction actions are cost effective modes in which the method of separation can be employed.

[0039] In a further aspect the invention relates to an apparatus including a drum having an axis, the drum being rotatable about a horizontal axis corresponding to the axis of the drum, wherein the drum is partially filled with particulate material having particle diameters in the range below 1 mm to around 1 μ m, the particulate material having a poly-dispersity of less than +/- 10%, wherein the particulate material has two boundary surfaces perpendicular to a central axis of the drum, preferably the particulate material occupies 10 to 90% of the filling volume of the drum.

[0040] In this connection it should be noted that the drum has a generally cylindrical shape, with a height perpendicular to a cylindrical base surface of the drum and the horizontal axis of the drum coincides with the central axis of the drum. In this connection it should be noted that the particulate material is introduced into the drum preferably from an end thereof.

[0041] Preferably the boundary surfaces of the particulate material are arranged adjacent either to particles having a larger diameter than the particulate material of interest or to a base or top surface of the drum having a surface roughness.

[0042] It is preferred when the particles having a larger diameter than the particulate material of interest are provided in the drum on each side of a region containing particulate material. The particles having a larger diameter are in particular arranged at each side of the region containing particulate material when the drum is arranged such that its central axis at least substantially coincides with the horizontal axis, this means that larger particles are arranged to the right and to the left of the particulate material wherein the drum is placed such that its central axis coincides with the horizontal axis in this example. Thereby the duration of a method of separating particulate material can advantageously be reduced, as an axial pre-segregation can be achieved in this way.

[0043] On a filling of the drum with particulate material a spacer element can be inserted along the jacket surface of the drum to ensure that the correct filling volume of the particulate material is achieved and that the particulate material is not prematurely mixed prior to a rotation of the drum about the horizontally arranged central axis.

[0044] The particulate materials can be introduced into the drum, e.g. from an end of the drum, in consecutive steps such that the particles having a larger diameter are e.g. arranged at each side of the region containing particulate material. This then looks like a sandwich structure of particles turned on its side. This means that the region containing particulate material is arranged in a middle region along a central axis of the drum without coming into contact with the end face of the drum. It is further preferred when a surface of the drum has a surface roughness.

[0045] In a further aspect the present invention relates to a particulate material having a poly-dispersity of less than 2% preferably of less than 1 % and most preferably of less than 0.1%, the particulate material is made using a method in accordance with the invention and/or by using an apparatus in accordance with the invention.

[0046] The invention will be explained in more detail in the following also with respect to further features and advantages by way of example with reference to embodiments and to the enclosed drawing. The Figures of the drawing show in:

Fig. 1a-c a scheme detailing the generation of axial and radial micro segregation,

Fig. 2 a representation of the axial and radial micro segregation of the particulate material,

Fig. 3 that a flux of small particles between two stripes is determined by both the stripe width ratio and the size of particles at the opposing stripe edges,

Fig. 4 coarsening control and microsegregation,

Fig. 5 an axial profile of the average diameter of the small grains in a stripe,

Fig. 6 micro segregation introduces an effective difference in bead diameter at the edges of adjacent bands of formally identical bead species,

Fig. 7 on use of small monodisperse particles long-term coarsening is absent,

Fig. 8 the obtained radial micro-segregation,

Fig. 9 a schematic representation of an apparatus in accordance with the invention,

Fig. 10 an agglomeration of particulate material in accordance with the invention,

Fig. 11 a cross section through a tomography demonstrating the initial step of the image processing, and

Fig. 12 radial distribution functions computed for different slices with minimum and maximum numbers of spheres.

[0047] Figs. 1 a to 1d show an apparatus 10 comprising a drum 12 for the segregation of a granular mixture 16 of

material comprising particulate material 20 having a poly-dispersity of substantially 10% and larger particles 18 in a horizontally rotating drum.

[0048] The drum 12 is a common device for the handling and processing of granular materials 16. The drum 12 is a partially-filled cylindrical container containing two sizes of grains 18, 20 which can rotate slowly around its horizontally-oriented axis C.

[0049] The drum 12 is further filled with a fluid 14 to compensate electrostatic effects between the grains of particles 18, 20. During rotation the particles 18, 20 are generally densely-packed and maintained in contact with one another. Only a thin layer of particles 18, 20 at the top roll and slide past one another other by means of so-called continuous flow dynamics during a horizontal rotation of the drum, i.e. the gravitational force on the particles on the top of the particulate material detach from the underlying material and move towards the bottom of the slop of the particulate material in the drum 12.

[0050] Fig. 1 a shows the state following which the granular mixture 16 and a fluid 14 have been introduced into the drum 12. The actual segregation of the granular mixture typically takes place in three stages.

[0051] The first stage is depicted in Fig. 1b in which radial segregation leads to the formation of a core region consisting of the smaller particles 20. This first stage is typically achieved following 15 to 20 rotations of a drum 12 having a diameter of 2 to 4 cm. This phase is well understood as the consequence of a sieving effect.

[0052] In many systems it is followed by axial segregation, this axial segregation typically arises after a further 50 to 500 rotations. This leads to the formation of a banded pattern as shown in Fig. 1 c.

[0053] Fig. 1d finally shows the coarsening of this pattern following a logarithmic decay of the number of axial bands with the number of rotations of the drum 12. This long-term dynamics happens on the timescale of thousands of rotations, it is clearly distinguished from a topologically similar merging of bands during the very early phase of axial segregation.

[0054] The process of coarsening can be attributed to the transport of small-sized beads through the core channel of small beads from one band of small grains to a neighboring band of small grains.

[0055] Fig. 2 shows the state in which axial and radial micro-segregation of the particulate material is achieved in one of the regions A shown in Figs. 1c and 1d. The particulate material 20 which initially has a poly-dispersity of less than $\pm 10\%$, is separated into axial and radial micro-segregated zones Z_1 to Z_n in which the particulate material respectively has a poly-dispersity of at least less than $\pm 2\%$, advantageously of as low as 0.1% .

[0056] According to the invention the particulate material 20 of interest is removed from at least one of these zones Z_1 to Z_n after a desired number of rotations. The desired number of rotations depends, on the one hand, on the radius of the drum used and, on the other hand, on the degree of poly-dispersity that is required. For a sufficiently large drum, i.e. a drum having a diameter of greater than 50 cm, a poly-dispersity of approximately 0.1% can be achieved in a time effective manner, which is commercially very viable.

[0057] The invention will now be described using the words of the inventors. The process of this exchange can be studied optically and using NMR imaging: For this purpose the mixer was initially prepared with two separate bands of small-sized beads, enclosed by bands of the large-sized grain component (cf. Fig. 3a). It was observed that the transport between two adjacent bands is unidirectional. The narrower band loses material to the larger one at a constant rate, until it is extinguished. Once this transport sets in, its direction is irreversible. The large beads do not contribute to the pattern dynamics. When a band of small beads separating two large-grained bands is extinguished, the latter merge to one region. If the experiment is continued for sufficiently long time (up to weeks), the result is complete segregation into one or two bands of large grains and another band plus the core channel containing the smaller grains (see e.g. Fig. 3d).

[0058] The agitation of the grains occurs by revolutions about the cylinder axis, while the directional transport is axial. It breaks the axial symmetry of the horizontal tube. Consequently one has to search for a mechanism that drives a unidirectional flow of the small grains, preferentially from smaller to broader segregation bands. The process reminds of a hydrodynamic situation where one container empties into a communicating vessel by pressure differences. Thus, one potential hypothesis for the driving mechanism of the flux could be an effective surface tension between the regions composed of large and small grains. Such a free surface energy could originate from an decrease in a configurational entropy at the interface.

[0059] In this study, it was demonstrated that two cooperative processes are responsible for the coarsening. First, it is shown that if two adjacent stripes contain grains of slightly different sizes near their edges, the transport of grains through the core channel is always from the edge containing smaller grains towards the edge containing larger grains. Secondly, a microsegregation mechanism inside the band of small beads is revealed: When the size distribution of the small beads is not exactly monodisperse, larger ones accumulate at the borders of the segregation stripes. The center of the stripes contains smaller grains.

[0060] These results have three important consequences: (1) they provide an explanation as to why directed transport proceeds from narrower to broader neighboring stripes containing initially the same grain compositions, (2) they allow to control the stability of individual stripes by adding a small amount of slightly larger grains, and (3) they lead to the prediction that bands of monodisperse small grains do not show merging or coarsening. We verify this prediction experimentally.

[0061] Fig. 3: shows the flux of small particles between two stripes is determined by both the stripe width ratio and the size of particles at the opposing stripe edges. **a)** image of a mixer with two prepared stripes of small particles embedded in large-size beads. Panels **b-e** show space-time plots of four scenarios. The central stripe of large beads (bright) is always 8 cm broad and separates two (darker) stripes of (S1) or (S2) particles (sizes see text). Top bars sketch the preparation conditions. **b)**

[0062] For same grain types, here S2, the narrower stripe (10.5 cm versus 14 cm) dissolves. **c)** If a stripe is prepared from the larger grains S2 (same widths as in b), the one with S1 grains dissolves. Panel **d)** demonstrates the dominance of grain diameter over stripe width (4 cm S2 versus 30 cm S1). **e)** Even a small amount (0.5 cm) of S2 beads on the inner edge of the left stripe with 7.5 cm S1 beads, triggers dissolution of the broad stripe (20 cm) with purely S1 beads.

[0063] The experiments were performed in a 66 cm long tube of 37 mm diameter, rotated at 20 revolutions per minute (not critical). The tubes are half filled with bands composed of large (1.62 ± 0.062 mm diameter in all experiments) and smaller glass spheres. The latter are obtained by sieving mixtures, we use two fractions with size distributions between 0.355 mm and 0.500 mm ('S1') and between 0.500 mm and 0.630 mm ('S2') [46]. The tube is filled up with water after preparation of the stripes (Fig. 3a). The interstitial fluid has only quantitative but no qualitative consequences for the coarsening process. We use water primarily to avoid static charging and to improve sample transparency. The tube is illuminated from the top and images are recorded automatically in intervals of 1 min. From long-term observations (between 15,000 and 200,000 rotations), we construct space-time plots of the tube axis profiles, in which regions with small beads S1, S2 appear darker than those of large beads.

[0064] Figure 3 shows four typical scenarios with differently prepared initial configurations. The plots show that (1) if two bands consist of the same particle types, the narrower one vanishes, see Fig. 3b, (2) if one band contains slightly smaller grains than the other, it vanishes even if it is broader, see Figs. 3c,d, and (3) it is sufficient that the band edge contains larger particles to stabilize it, see Fig. 3e. This allows the stabilization of a dissolving band by adding a few S2 grains (Fig. 4, left): After a narrow band of S1 grains had already lost half of its content to the neighboring S1 band, we added $\approx 15\%$ of S2 material to it. This led to the immediate reversal of the transport through the core channel, even though the competing band contained almost 3 times as much material.

[0065] Fig. 4: shows coarsening control and microsegregation. Left: After the narrower of two prepared pure S1 stripes starts to dissolve, we add an small amount S2 material (equivalent to 1 cm band) to the dissolving 6.5 cm broad S1 band (circled region). This reverses the transport and the 17.5 cm broad S1 stripe starts to decay immediately. Right: Stripes of smaller white (S1') and larger red beads (S2', see text) beads were prepared. A core quickly forms from material of both stripes (thus, the regions of large grains also adopt a pink appearance). The S1' band transfers its material to the S2' band, where it is not simply deposited at the edge but forms a clearly segregated central band, surrounded by S2' beads.

[0066] The second process active in the coarsening is microsegregation inside each individual band of small particles. It moves the S2 beads in a mixed S1/S2 region to the band edges where they effectively control the material flow. Even when the band grows by incoming S1 grains, these are transported into the center of the stripe and the edge region remains occupied by the larger S2 grains. This is demonstrated with small colored (S2', 0.63-0.71 mm) and transparent (S1', 0.5-0.63 mm) spheres in Fig. 4, right. Initially, two bands were prepared where one contained only S1', the other one contained only the S2' fraction. The S1' material is transported into the growing stripe and in there, microsegregation places the S1' fraction in the stripe center. The edges between both S1' and S2' bands are astonishingly sharp, even though both species are neighboring sieving fractions.

Most importantly, however, microsegregation also occurs in stripes of particles with a much narrower size distribution. This can be verified by X-ray tomography in a down-sized system (tube diameter 24 mm, length 60 mm) filled with beads of diameters 1.01 ± 0.01 mm and 423 ± 23 μm (roughly Gaussian diameter distribution, measured with a P4 Retsch Camsizer (as was available at the time of filing this application under <http://www.retsch-technology.com/rt/products/dynamic-image-analysis/camsizer-p4/versions-accessories/>).

[0067] Measurements were performed in a Nanotom (GE Sensing and Inspection) with 40 μm voxel size $\ddagger$. A detailed description of how tomographic images of this resolution can be used to determine the average particle diameter d_{avg} with an accuracy better than a μm will be given at the end of the description.

After detection of the individual small and large particles the average diameter as a function of the axial coordinate was measured. The analysis of the small particle diameter in homogeneously prepared single stripes of small grains shows that beads are redistributed and micro segregated by the rotation of the mixer. Figure 5 shows the axial profile of the average diameter of the small grains in such a stripe. Initially, the mixture is uniform (black, horizontal curve). After 1,000 rotations of the mixer, a stable micro segregation is established and beads near the edges of the stripe are on average 2 % larger than those in the stripe center. As demonstrated below in Sec. 3.4, we observe radial micro segregation as well. However, this effect is not related to the flow through the core channel, thus we focus here on the axial micro segregation only.

Fig. 5: shows the axial micro segregation inside a single stripe of small beads (423 ± 23 μm) surrounded by larger beads. The average diameter in dependence on the axial positions is measured using x-ray tomograms. Symbols indicate

a bin size of 1.64 mm (●), 4.04 mm (○), and 8.44 mm (v). There is a clear tendency for the larger grains to accumulate at the band edges.

[0068] It is this axial microsegregation that is responsible also for the extinction of the narrower of two neighboring bands of beads with nominally the same diameters (c.f. figure 3 b). This is demonstrated with the experiment shown in figure 6. Using again X-ray tomography, it is shown that in a rotating drum prepared with two stripes of the same small particles, two combined features occur, namely the narrower stripe dissolves *and* the dissolving stripe displays a smaller mean bead diameter at its boundaries. This once more confirms that the direction of the flux between is pointing from the stripe containing smaller particles to the one with larger particles.

[0069] Two considerations complete this argument. First, the smaller mean particle diameter at the boundary of the narrower stripe can be understood as a consequence of the smaller reservoir inside its bulk which cannot provide the same amount of largest particles as they are present in the broader stripe. Second, this mechanism is self-sustaining. Once the flow from the narrower stripe to the broader sets in, the narrower stripe preferentially loses its largest particles which had accumulated at the boundary.

[0070] Fig. 6: shows that micro segregation introduces an effective difference in bead diameter at the edges of adjacent bands of formally identical bead species; here $423 \pm 23 \mu\text{m}$. All data are measured using X-ray tomography. The vertical lines mark the stripe boundaries (equal volumes of small and large spheres). Symbols indicate the bin size used for the

diameter measurements: 1.64 mm (●), 4.04 mm (○), and 8.44 mm (v)

[0071] The coarsening appears to be absent in systems where the small particles are monodisperse. Thus, drums with mixtures of large-size beads of $2.62 \pm 0.065 \text{ mm}$ and monodisperse small spheres of diameter $1.01 \pm 0.01 \text{ mm}$ were prepared. As shown in Fig. 7, the initially prepared bands remain either stable or start to drift or jitter. However, there is no coarsening on the timescale of the experiments, an order of magnitude longer than the experiment displayed in Fig. 3 b). The positional jitter of the thin stripe in Fig. 7b is an indication that the shown experimental timescales are sufficient, the slow exchange of large beads tunneling through the thin stripes becomes comparable to the small particle dynamics.

At this point the crucial question is, why the transport in the core channel between two stripes is related to the particle size differences at the interfaces. In the following two hypotheses are proposed: the driving force could either be an effective surface energy which depends on the diameter ratio at the interface between small and large particles. Alternatively, a gradient in small particle concentration could determine the drift. Either hypothesis requires further experimental verification. Our findings agree well with observations of other researchers.

Figure 7: shows that long-term coarsening is absent when the small particles are monodisperse. Systems were prepared with two small bands composed of monodisperse small glass spheres ($1.01 \pm 0.01 \text{ mm}$) surrounded by larger ($2.62 \pm 0.065 \text{ mm}$) glass spheres. Left: Space-time diagram of the evolution of two stripes of equal initial size. Only drift but no coarsening is observed. Right: Two bands prepared with different initial widths, 5 cm and 8 cm, resp. Again no coarsening (but a pronounced lateral jitter) was observed.

[0072] Microsegregation is also present in radial direction (see also Fig. 2 in which the zones Z_1 to Z_n of micro-segregated particles have both a radial and an axial extent.

Figure 8 shows that the mechanism establishes, especially in the center of the stripe, a gradient from smaller to larger diameters when moving outwards from the core (see also the Zones Z_1 to Z_n shown in Fig. 2). However, as this effect does not break axial symmetry it is not related to the uni-directional flow behind the long-term coarsening phenomenon. One possible explanation is the existence of an effective free surface energy σ at the interface of each stripe where σ increases with the difference between small and large particle diameters $d_l - d_s$. It is always the stripe with the larger ($d_l - d_s$) and therefore the larger σ which dissolves. In order to explain the sequence of stripes evolving in Fig. 4, one needs to assume that σ depends on $(d_l - d_s)^\alpha$ with an exponent α larger one. An argument for the origin of σ can be derived from the assumption of an Edwards ensemble where a configurational entropy S_{conf} is defined as the logarithm of the number of possible mechanically stable configurations for the given volume fraction and boundary stresses. The important point is that the number of possible packings of monodisperse spheres does not depend on their diameter. Therefore in the bulk of both large and small stripes, the entropy per particle S_{conf}/N^* is approximately identical. However, at an interface between two sizes of particles, the number of mechanically stable configurations will depend on the diameter ratio. This could provide an effective surface tension $\sigma(d_l - d_s)$. The second hypothesis presupposes that the linking channel contains a concentration gradient dc_s/dx of small beads that drives their directed motion.

[0073] Fig. 8 shows radial microsegregation. This figure shows data taken after 1000 rotations. The three kinds of data points describe three radial bins where the distance is measured from the outer surface of the sample. The blue

data marked as $> 4 \text{ mm}$ corresponds to the inner core of the sample. Symbols indicate axial bin sizes of 1.64 mm (●), 4.04 mm (○), and 8.44 mm (v). Bins contain at least 2000 (0 - 1.8 mm and 1.8-4 mm), respectively 1000 ($> 4 \text{ mm}$) small spheres.

[0074] This has been established experimentally by varying the size ratios of large and small beads that the equilibrium concentration of the smaller species in the core channel, c_s (number of particles in the cross section) grows with the size ratio $r = d/l/d_s$. Different r at the interfaces of two neighboring bands of small beads (caused by different compositions of the stripe edges) will therefore lead to a concentration gradient, c_s becomes larger at the end where r is larger (narrower segregation band or S1 beads). An effective particle flow towards the opposite channel end (large segregation band or S2 beads) is the consequence, until the band which provides the particles is completely extinguished.

Two mechanisms are revealed in this work that govern the coarsening of axially banded pattern of bi-disperse granular mixtures in a horizontally rotating drum: micro-segregation accumulates larger particles of the small component at the band edges, and material transport generally sets in from bands with smaller particles at their edges to neighbors containing larger particles at the edge. When the small-sized beads are monodisperse, no coarsening of the pattern occurs, even over long experimental timescales. We demonstrated that the stripe stability can thus be controlled by addition of a few percent of different-sized grains. Furthermore, the observed micro-segregation has been described here for the first time. It might be exploitable in technological processes where particles are to be sorted efficiently by size. Fig. 9 shows a schematic representation of an apparatus 10 in accordance with the invention. The drum 12 has a generally cylindrical shape with the central axis c of the drum being arranged at least substantially horizontally. The drum 12 is filled with a fluid 14, particulate material 20 with a poly-dispersity of less than $\pm 10\%$, and with larger particles 18. The fluid 14 is present at the top of the horizontally arranged drum 12, whereas the larger particles 18 are arranged at either side of the particulate material 20.

[0075] Typical drum sizes that are suitable for carrying out the method in accordance with the invention have radii ranging from 2 cm to 200 cm and lengths ranging from 30 cm to 300 cm. For shorter but wider drums one Region A (see Figs. 1 c and 1 d) can be achieved, whereas for longer tubes a plurality of Regions A can be achieved. Each Region A then contains several Zones Z1 to Zn in which the poly-dispersity of the particulate material 20 is less than that of the initial poly-dispersity, the number of zones depends on the number of rotations of the drum, which in turn also depends on the size of the drum selected and on the resolution of the individual zones required for the specific application.

[0076] Fig. 10 shows an agglomeration of particulate material 20, wherein the particles of the particulate material have a poly-dispersity of substantially less than 0.1%. Such a poly-dispersity can be achieved by rotating the drum about the horizontal axis approximately 1500 times for a given material and a given particulate material size.

[0077] In order to process the data obtained using x-ray tomograms a series of steps is carried out.

[0078] The initial step is a precise determination of the centers of all small and large particles. This task is performed with a suite of matlab programs which first determine the centers of all large particles by finding the maxima in a convolution of the tomography with a template of a large spheres. Then all large particles are removed from the tomography by setting the corresponding voxels to black (c.f. Fig. 11).

In the next step all small spheres are detected again with voxel resolution by a second convolution with a small sphere template. Finally, the position of the small particles is measured with sub-voxel accuracy using an interpolation of the convolution results in the direct neighborhood of the maximum.

The second step is the computation of radial distribution function $g(r)$ which gives the probability to find the center of a neighboring sphere within a shell of radius $[r; r+\Delta r]$. In our analysis we approximate $g(r)$ by using shells with a width $\Delta r = 2 \text{ m}$ i.e. 0.05 voxel. Fig. 11 gives two examples.

The final step is based on the fact that the first peak of the radial distribution function, i.e. the most likely distance between two particles, corresponds to them touching. Therefore determining the position of the first peak of $g(r)$ with a Gaussian fit is also a way to measure the average diameter d_{avg} of all particles. These fits were performed using the following:

$$g(r) = a \cdot \exp \left(-\frac{(r - d_{avg})^2}{2\sigma^2} \right) \quad (1)$$

[0079] in an interval of $\pm 0.05 \text{ mm}$ around the approximate maximum determined by lowpass filtering of $g(r)$.

In Figs. 5 and 6 the evolution of d_{avg} along the center x-axis of the cylinder is shown by computing $g(r)$ for all small spheres within bins of size $x \pm w/2$. However, in order to have a sufficient statistic the window width w has to be adapted in order to guarantee that there are at least 2000 small particles within each window. The two graphs in Fig. 12 correspond to bins with the minimal and maximal number of small spheres included in the computation of $g(r)$.

[0080] Fig. 12 shows the radial distribution functions computed for different slices with minimum and maximum numbers of spheres in Fig. 5. Left: slice of width $w = 1.64 \text{ mm}$ at the axial position of $x = 16 \text{ mm}$. This slice contained 5606 small spheres. Right: slice of width $w = 8.44 \text{ mm}$ at the axial position of $x = 32 \text{ mm}$. The lines are fits to the peaks using Eq. (1).

Claims

1. A method of separating particulate material with particle diameters in the range below 1 mm to around 1 μ m, the particulate material having an initial poly-dispersity of less than +/- 10%, the method including the steps of:
 - a) inserting the particulate material into a drum having an axis so that the drum is partially filled with the material,
 - b) rotating the drum with the particulate material therein about its axis, with the axis of the drum being generally horizontal, at a rate such that a free surface of the particulate material shows continuous flow dynamics,
 - c) continuing the rotation at least until radial micro-segregation of the particulate material is achieved, and
 - d) extracting particles from at least one zone of the micro-segregated particles having a poly-dispersity of substantially less than the initial poly-dispersity.
2. A method in accordance with claim 1, wherein the particulate material comprises spherical particles.
3. A method in accordance with claim 1 or 2, wherein the particulate material comprises particles having the same density.
4. A method in accordance with at least one of the preceding claims, wherein the partially filled drum is additionally filled with a fluid.
5. A method in accordance with claim 4, wherein the fluid is a gas or a liquid that is selected from the group comprising liquids capable of freezing, liquid phases of alcohols, liquids capable of easily vaporizing, liquid waxes, as well as liquids which permit the dissipation and screening of electrostatic charges.
6. A method in accordance with any one of the preceding claims, wherein a drum is partially filled with particulate material when the particulate material takes up 10 to 90% of a filling volume of the drum.
7. A method in accordance with any one of the preceding claims, wherein the speed of rotation of the drum is selected below the threshold at which centrifugal effects modify a distribution of the particulate material present in the at least one zone of the micro-segregated particles.
8. A method in accordance with any one of the preceding claims, wherein particles having a larger diameter than the particulate material of interest are further provided in the drum.
9. A method in accordance with any one of the preceding claims, wherein a surface of the drum is provided with a surface roughness to enhance at least the radial micro-segregation.
10. A method in accordance with any one of the preceding claims, wherein the poly-dispersity of the particulate material in the at least one zone of the micro-segregated particles is less than 2%.
11. A method in accordance with any one of the preceding claims, wherein particles having a desired poly-dispersity are continuously or discontinuously extracted from the at least one zone.
12. A method in accordance with claim 11, wherein the continuous or discontinuous process comprises a scraping or a suction action.
13. An apparatus, preferably for carrying out the method of any one of the preceding claims, including a drum having an axis, the drum being rotatable about a horizontal axis corresponding to the axis of the drum, wherein the drum is partially filled with particulate material having particle diameters in the range below 1 mm to around 1 μ m, the particulate material having a poly-dispersity of less than +/- 10%, wherein the particulate material has two boundary surfaces perpendicular to a central axis of the drum.
14. An apparatus in accordance with claim 13, wherein the boundary surfaces of the particulate material are arranged adjacent to either particles having a larger diameter than the particulate material of interest or to a base or top surface of the drum having a surface roughness.
15. A particulate material having a poly-dispersity of less than 2% and made by any one of the preceding method claims 1 to 12 and/or using an apparatus in accordance with claim 13 or claim 14.

Fig. 1a

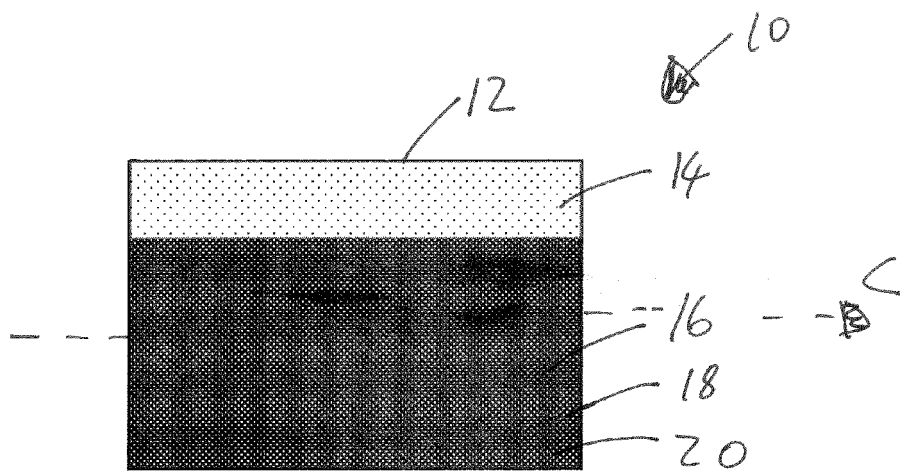


Fig. 1b

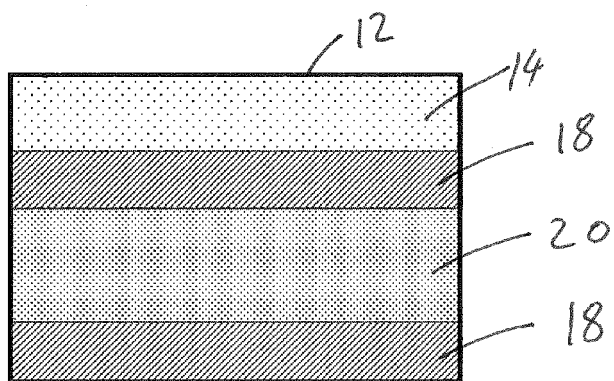


Fig. 1c

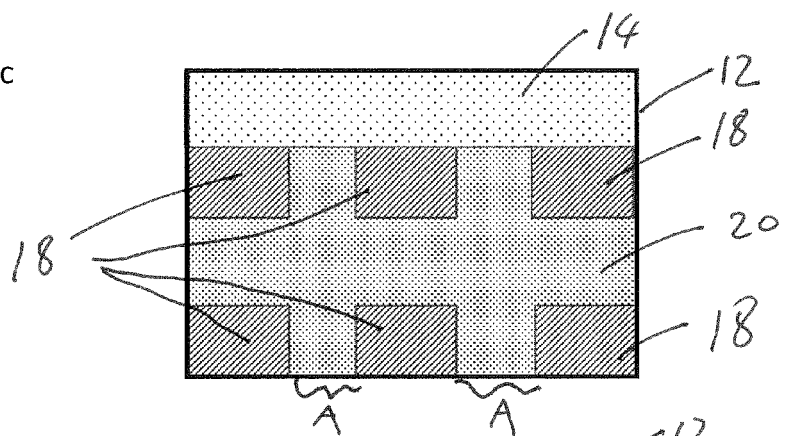


Fig. 1d

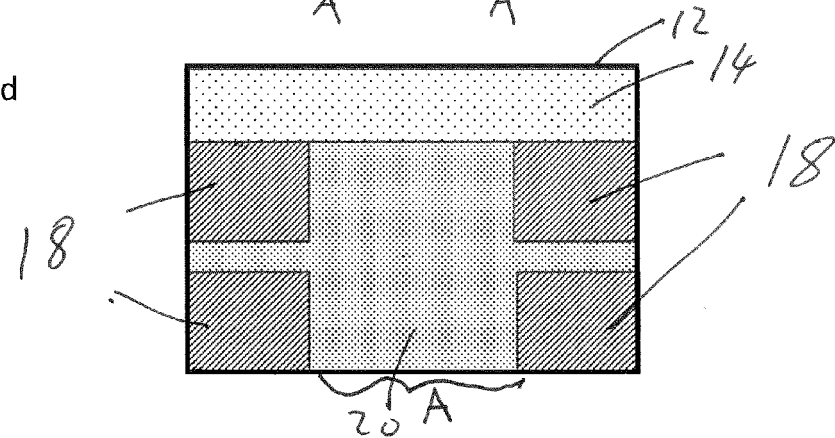


Fig. 2

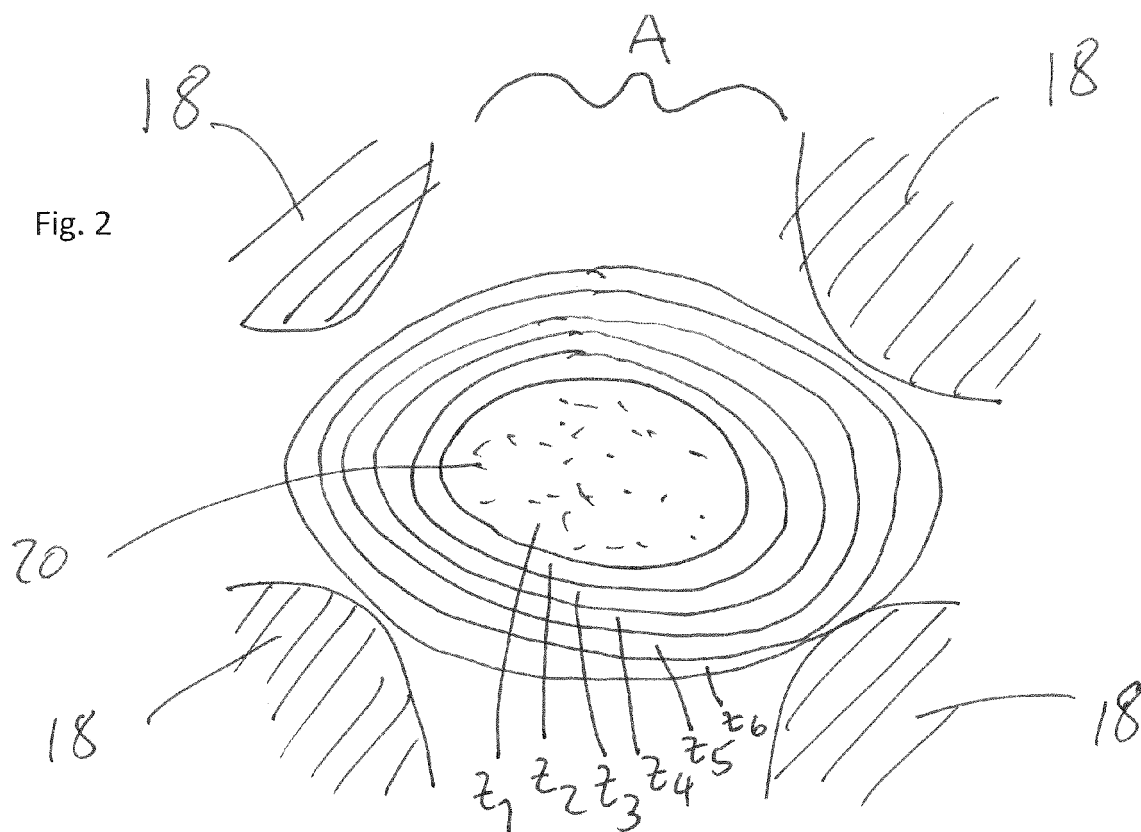


Fig. 3

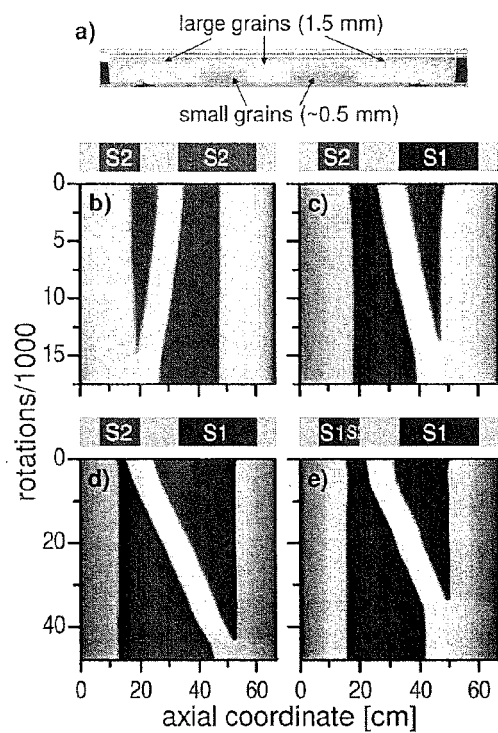


Fig. 4

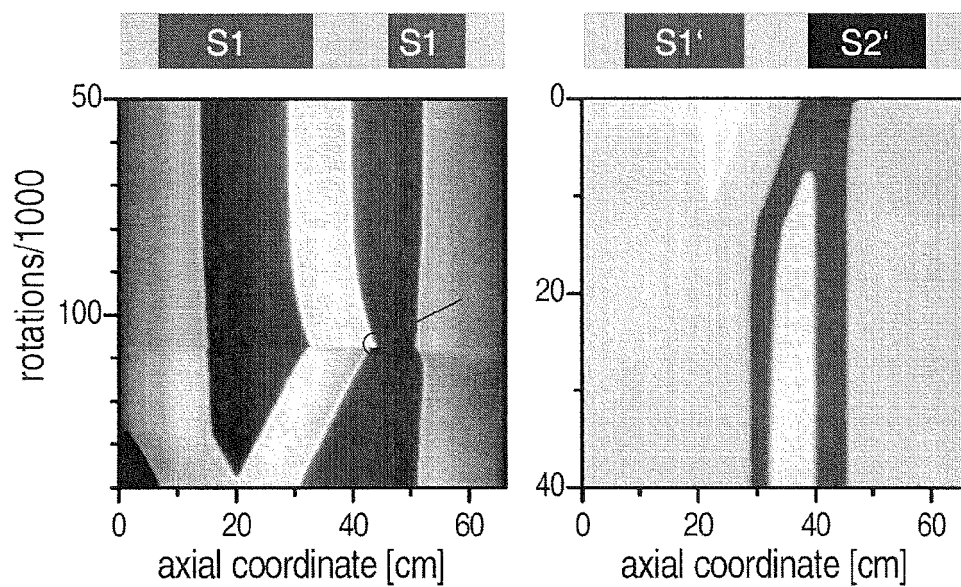


Fig. 5

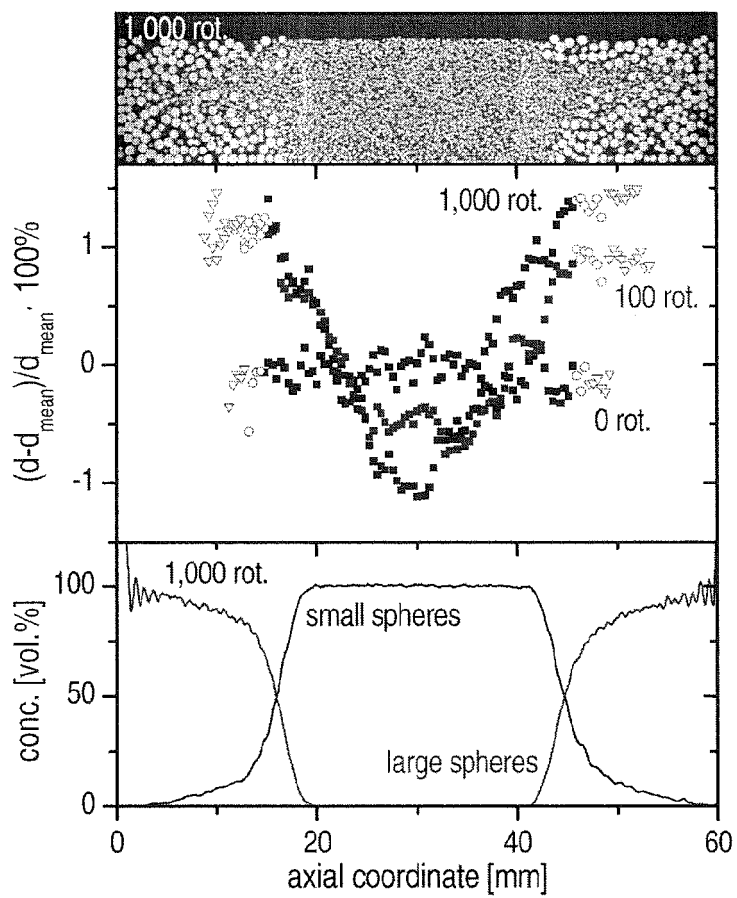


Fig. 6

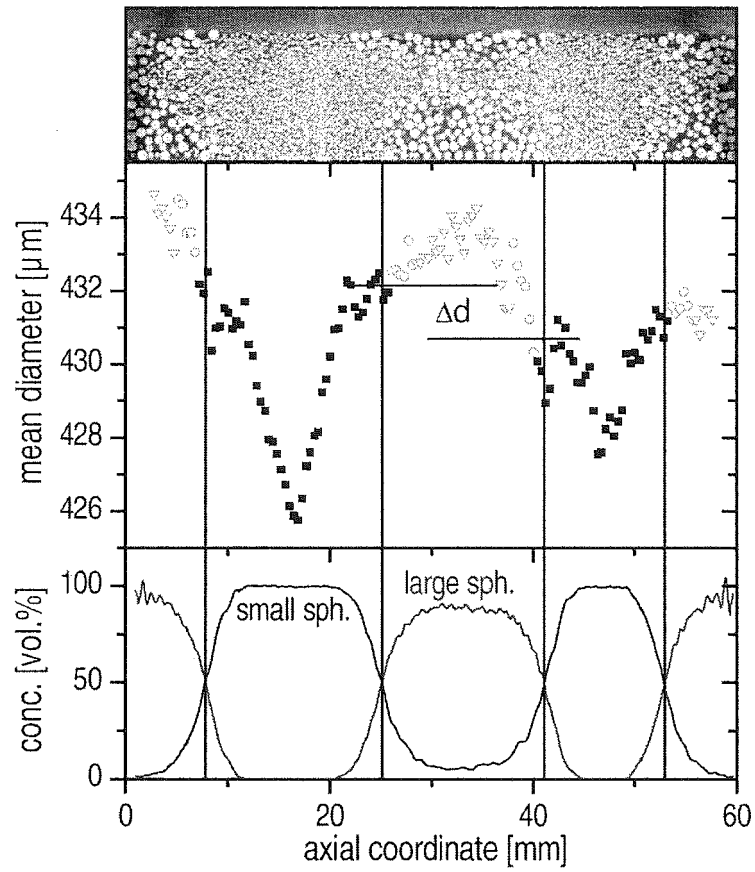


Fig. 7

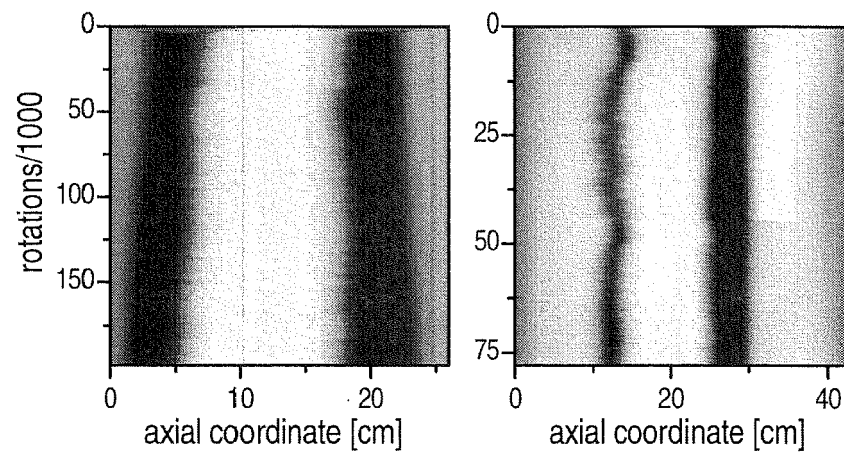


Fig. 8

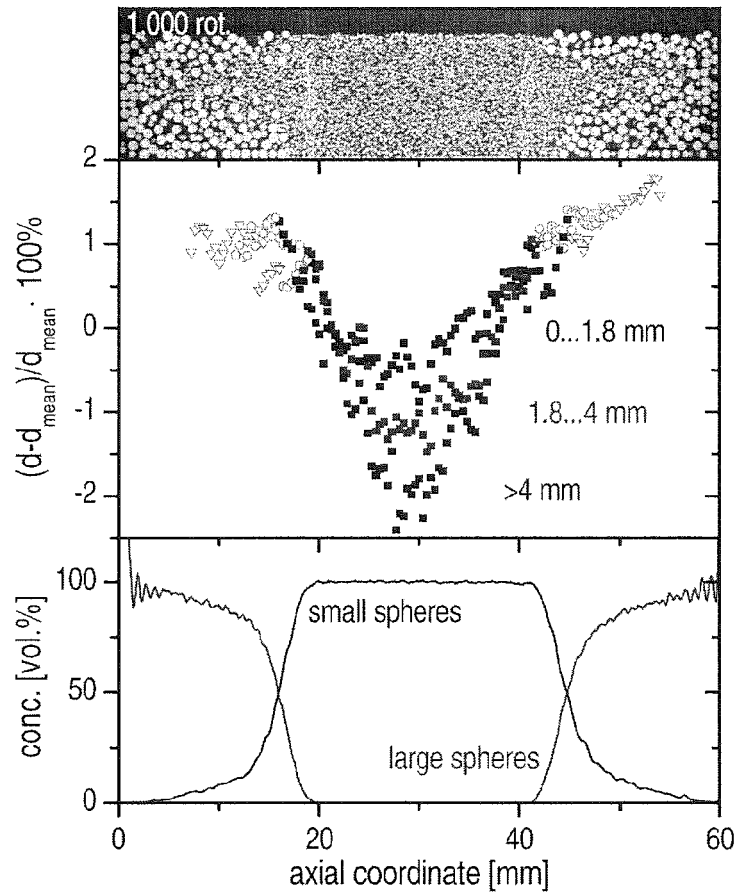


Fig. 9

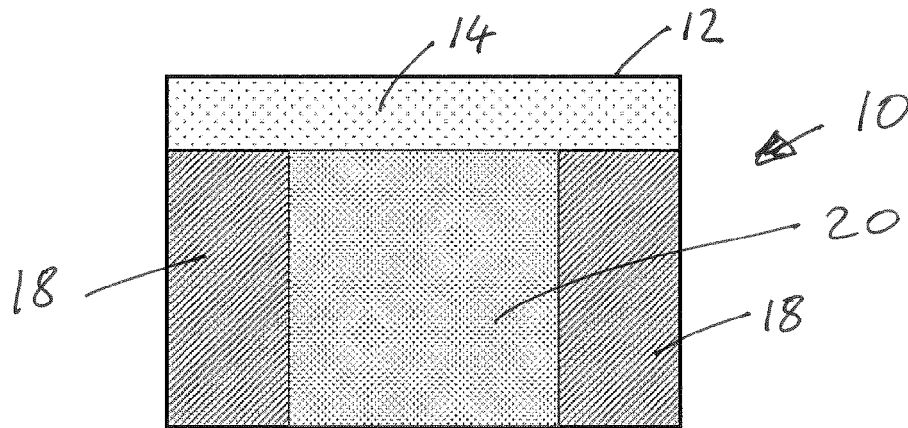


Fig. 10

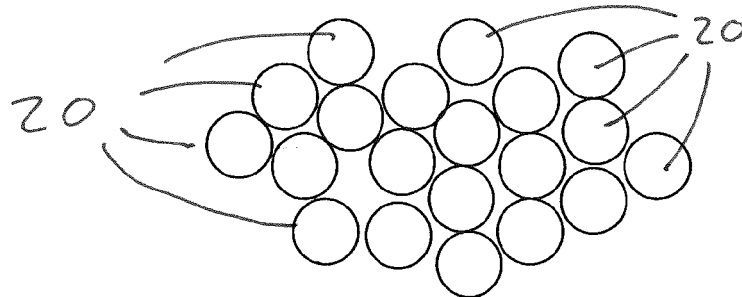


Fig. 11

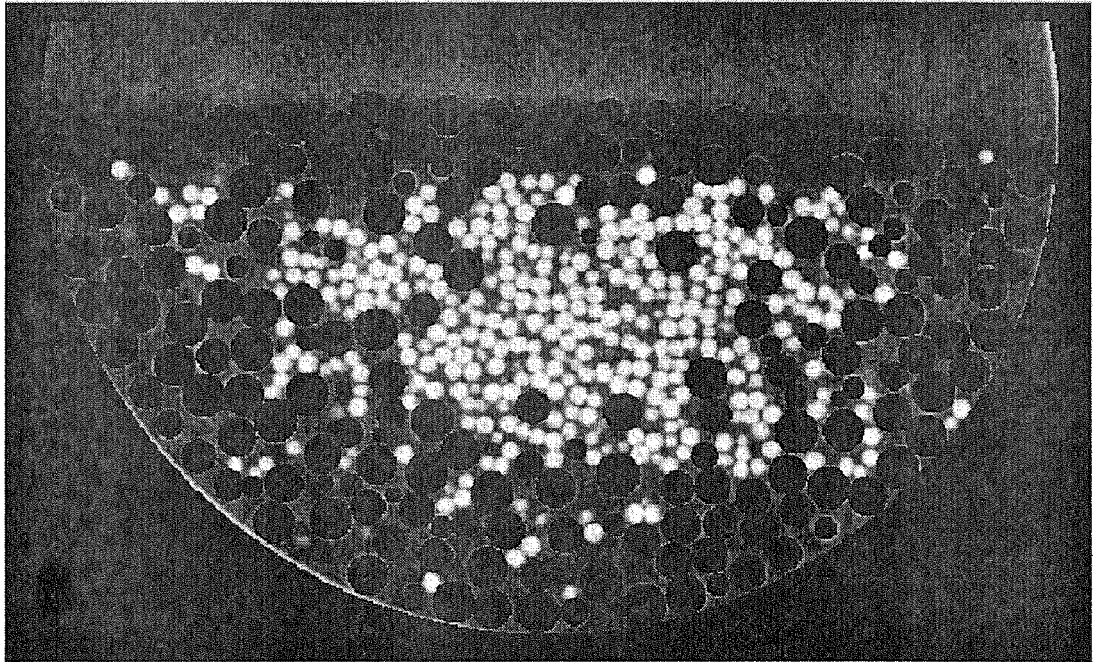
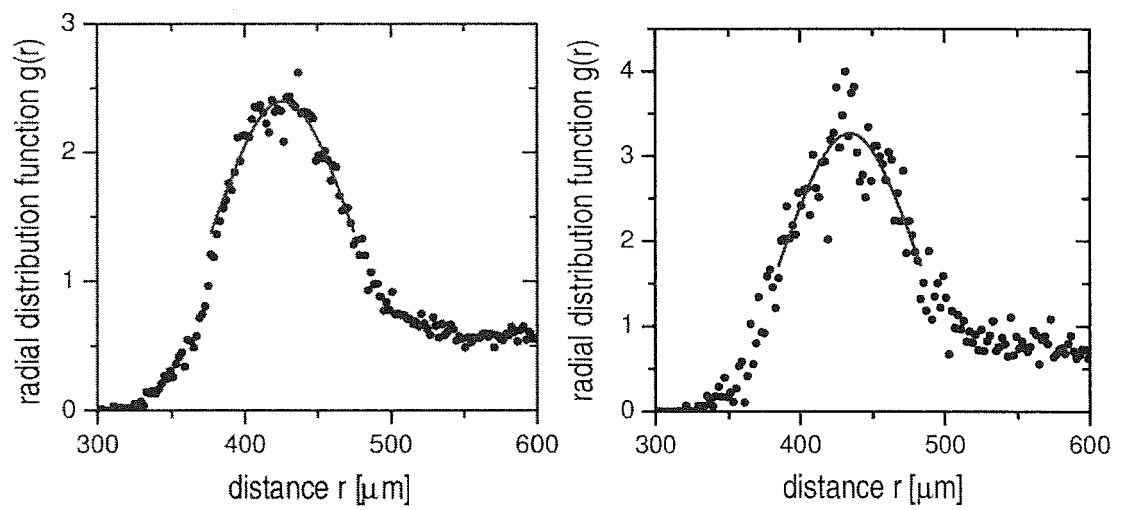


Fig. 12





EUROPEAN SEARCH REPORT

 Application Number
 EP 15 18 0048

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DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
X	Tilo Finger ET AL: "Granular materials", Physical review. E, Statistical, nonlinear, and soft matter physics, 21 February 2013 (2013-02-21), XP055239634, United States DOI: 10.1103/PhysRevE.80.061302 Retrieved from the Internet: URL:http://www.uni-magdeburg.de/anp/projects/granular/granular2013.html [retrieved on 2016-01-08]	13-15	INV. B03B5/56
A	* page 1 - page 2 *	1-12	
X	TILO FINGER ET AL: "Influences of the interstitial liquid on segregation patterns of granular slurries in a rotating drum", PHYSICAL REVIEW E (STATISTICAL, NONLINEAR, AND SOFT MATTER PHYSICS), vol. 75, no. 3, March 2007 (2007-03), XP055239633, US ISSN: 1539-3755, DOI: 10.1103/PhysRevE.75.031308	13-15	
A	* page 1, paragraph 1 - page 3, left-hand column, paragraph 1; figures *	1-12	
The present search report has been drawn up for all claims			
Place of search Munich		Date of completion of the search 11 January 2016	Examiner Leitner, Josef
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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