

Reversible axial segregation of binary mixtures of granular materials

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Measurements of axial segregation of binary mixtures of glass beads having different diameters in a rotating horizontal cylinder are reported. For rotation speeds of ~ 15 rpm a mixed state quickly (within a few minutes) segregates into sharp bands of alternating large and small glass beads along the axis of rotation. Decreasing the rotation speed to ~ 5 rpm causes the bands to disappear, restoring the mixed state. This process is reversible upon increasing the rotation rate, though the exact location and width of the bands vary from run to run. The axial segregation phenomena are analyzed in terms of a diffusion equation where the effective diffusion coefficient is the difference between segregating drift and normal Fickian diffusion.

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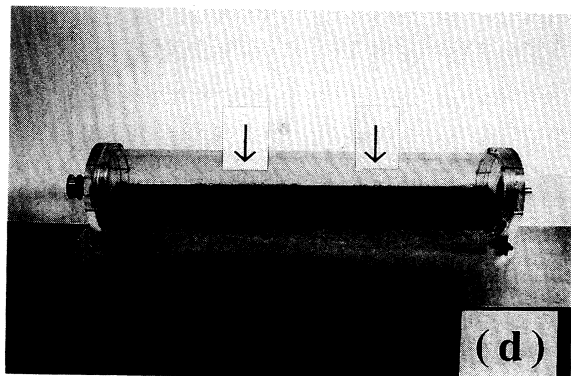
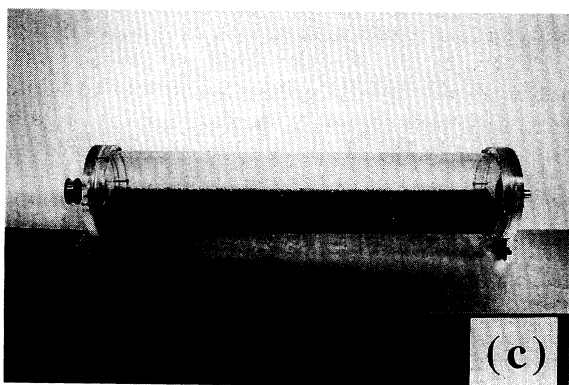
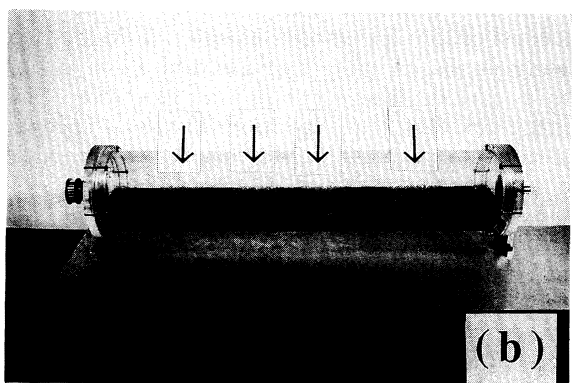
A fascinating property of binary mixtures of granular media such as sand and powders is the phenomenon of mass or size segregation when the mixture is shaken or rotated [1–10]. When a horizontal cylinder is partially filled with a mixture of two different types of granular media, such as different sizes or masses of glass beads or sand, and then rotated about its long axis, the binary mixture segregates into alternating bands of relatively pure single concentrations along the axis of rotation [3–10]. One proposed explanation of this phenomenon, termed axial segregation, involves a diffusion equation for the relative concentrations of the individual species in the mixture, where the ordinary diffusion coefficient is replaced by an effective diffusion coefficient which depends on the relative contributions of normal Fickian diffusion (tending to mix the two species) and a preferential drift term [9]. The drift term arises from a difference between the dynamic angle of repose of the mixed and segregated states. When the drift term is larger than the Fickian diffusion term, fluctuations of separated species grow with time.

The phenomenon of axial segregation is well known in the engineering literature and has recently attracted attention as an additional mechanism, other than vertical shaking, by which binary granular mixtures can spatially segregate. While segregation of binary mixtures of granular media, either due to shaking or rotation, is well known, there has been to date no description of a reversible segregation phenomenon. In this paper we report experimental measurements of axial segregation for a binary mixture of glass beads for which the segregated state can be reversed, restoring the mixed state, simply by changing the speed of rotation. Measurements of the dynamic angle of repose ϕ as a function of the rotation speed confirms that the difference between ϕ of the mixed and segregated phases varies with the rotation speed, in agreement with our observations of mixing and demixing phenomena.

A basis for a statistical mechanics of powders was recently described by Edwards and Mehta [11]. While in a thermodynamic system the energy of the system depends on the configurations of the constituent microscopic particles, for a granular material it is the volume rather than the energy which depends on the configurations of the

individual particles. Using the standard definition of entropy, they defined the compactivity X of the granular material as $X = \partial V / \partial S$, which fulfills the same role as temperature in conventional statistical mechanics. For a vertically vibrated binary mixture of different size sand particles they were able to predict the occurrence of size segregation as the compactivity is varied to a critical value X_c . However, thermodynamic phase transitions are reversible when the temperature is cycled through the transition point. While an analysis of the compactivity for the axial segregation phenomenon is not available, it seems reasonable that as the rotation speed is varied there will be a corresponding change in the compactivity of the granular media. The reversible axial segregation phenomenon described here may therefore serve as a model system for studying phase transitions and the statistical mechanics of granular media.

The experimental setup (similar to that employed in Ref. [8]) consists of a Plexiglas tube 12.7 cm in diameter and 2 ft long (identical results are obtained with a cylinder 3 ft long) with Plexiglas end plates bolted onto both ends of the cylinder. Brass flanges on each end plate contain bolts coaxial with the center of the cylinder which reside upon rotating ball bearing mounts. One of the bolts has an additional flange at its end, which is connected to a 1/17 hp motor. The sand used in the study is actually glass beads of approximately uniform size with diameters 2.85 mm (termed “large” beads) and 0.5–0.75 mm (termed “small” beads). The cylinder is filled halfway with sand composed of a mixture of 50% by volume of large and small glass beads, which are either premixed or mixed in the cylinder by manual shaking and rocking, as shown in Fig. 1(a). The smaller beads are dyed blue to improve the contrast in the photographs; identical results were observed with undyed beads. The horizontal cylinder is leveled using shims, though the results do not depend on precise leveling. After rotation for 13 min at a rotational speed ω of 14 rpm, clear axial segregation of the small and large beads into alternating bands is evident, as shown in Fig. 1(b). When the motor speed is decreased to 5 rpm for 1 h, the bands disappear and a homogeneous mixture is restored [Fig. 1(c)]. Finally upon increasing the motor speed to $\omega = 14$ rpm for 13



min, the bands reform as shown in Fig. 1(d), though in slightly different locations and with different widths.

While the location and widths of the bands in Figs. 1(b) and 1(d) vary from run to run, there are three features of the axial segregation which are highly reproducible. First, almost immediately upon rotation there is segregation of small beads near the radial origin of the cylinder along its entire length. This radial segregation has been reported previously [4–6], and is most likely a manifestation of size segregation due to the variation in compactivity below the avalanching surface in the rotating drum [11]. The second reproducible feature is that the first regions to undergo axial segregation are directly adjacent to the end plates, and large beads always form narrow bands there before the rest of the mixture segregates [6]. This is observed in both the 2 ft and 3 ft long cylinders and is clearly a boundary effect. The third reproducible feature is that the bands in Figs. 1(b) and 1(d) are not stable, but for longer rotation times at high speeds these bands eventually always merge into one central band of large beads with bands of small beads adjacent on either side (with the narrow bands of large beads at the end plates). The time dependence of the band formation process will be described in a later publication [12].

Axial segregation is also observed for a 50-50 mixture of “medium” sized glass beads (diameter ~ 1.2 mm) and sand (diameter ~ 250 – 500 μm) and for the small glass beads of Fig. 1 mixed with sand. However, neither of these binary mixtures of granular media exhibits a remixing of the segregated bands upon changing the rotation speed. As discussed below, a reversible axial segregation effect is seen when the difference of the dynamic angle of repose between the mixed and segregated phases is zero at finite rotational speeds. This condition applies for the system in Fig. 1 but not for the other mixtures which show a nonreversible segregation effect. We note parenthetically that smooth round beads are not necessary for this phenomenon; we have observed axial segregation of a mixture of split peas and uncooked rice. After rotation for approximately 2 h at 14 rpm a central band of peas forms with two side bands of rice. A uniform rotation speed is also not necessary; we have achieved axial segregation when the cylinder was disengaged from the motor and rapidly accelerated by hand.

We now address the physical mechanism responsible

FIG. 1. Photographs of the horizontal rotating cylinder described in the text. The cylinder is filled halfway with a 50-50 mixture by volume of large glass beads (average diameter 2.85 mm) and small glass beads (average diameter 0.5–0.75 mm). Photo (a) shows the initial setup, with the glass beads homogeneously mixed. After rotation about the long horizontal axis at 14 rpm for 13 min, axial segregation of the large and small glass beads is evident [photo (b)]. Further rotation for 1 h at 5 rpm remixes the two phases back into a homogeneous mixture, as shown in photo (c). Finally, the axial segregated state is restored by additional rotation at 14 rpm for 13 min [photo (d)].

for the axial segregation phenomenon, following arguments proposed by de Gennes and Savage [9]. If the two different granular media have different dynamic angles of repose ϕ , then a drift current will develop along the axis of rotation which tends to separate the two species [4–7,9,10]. As pointed out by Bridgwater, Sharpe, and Stocker [5], when the mixed phase has a greater angle of repose than the separated phases a small segregated fluctuation will tend to grow, leading to a full separation of the two species. Whichever of the two phases has the lower dynamic angle of repose will segregate out of the mixed phase first; when the mixed phase is fully depleted of this species it will also be a segregated band of the other granular material. As indicated schematically in Fig. 2, the height of the sand mixture z will vary along the axis of rotation x due to this angular difference. Hence further small perturbations will tend to send additional mass out of the mixed phase into the adjacent segregated band (assumed here for sake of argument to consist of the large glass beads). This drift current j_{drift} out of the mixed phase will be proportional to the gradient of the large bead concentration C_l and can be written as

$$j_{\text{drift}} = \beta \partial C_l / \partial x . \quad (1)$$

Here β is a proportionality constant which is a function of the height variation along the x axis, and can be expressed as a monotonic function of the difference between the dynamic angles of repose (in particular, when $\Delta\phi = 0$ then $\beta = 0$). Due to random collisions between the glass beads flowing down the avalanching surface there will also be the traditional Fickian diffusion, which will mix together the two segregated bands. This diffusion current j_{diff} is given by the expression

$$j_{\text{diff}} = D \partial C_l / \partial x , \quad (2)$$

where D is the diffusion coefficient for the large glass beads (for simplicity we assume that the small and large beads have the same D , though this is not crucial). The net current is clearly $j_{\text{tot}} = j_{\text{drift}} - j_{\text{diff}}$ and the rate of growth of the segregated phase C_l is given by the continuity equation

$$\frac{\partial C_l}{\partial t} = -\frac{\partial}{\partial x} j_{\text{tot}} = -\frac{\partial}{\partial x} [(\beta - D) \partial C_l / \partial x] , \quad (3)$$

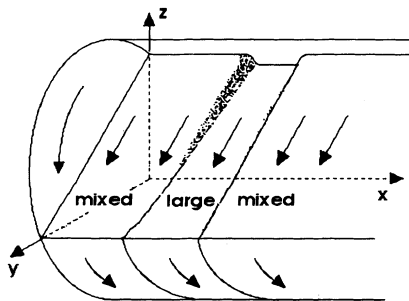


FIG. 2. Schematic diagram indicating the higher dynamic angle of repose for the mixed state compared to an adjacent narrow band of segregated large glass beads.

which resembles a standard diffusion equation if we replace $D - \beta$ with D_{eff} . However, when $\beta > D$, then $D_{\text{eff}} < 0$ and one has a diffusion equation with the unusual property that collisions tend to order (that is phase segregate) rather than further mix two species [9]. While providing a phenomenological justification for axial segregation, this simple analysis has ignored the fact that the avalanching surface actually consists of diffusing beads approximately eight particle diameters deep below the free surface. Since β depends on $\Delta\phi$ which varies along the x axis, β is a function of x as well, and Eq. (3) can also be expressed as

$$\partial C_l / \partial t = -(\partial\beta/\partial x)(\partial C_l / \partial x) + D_{\text{eff}} \partial^2 C_l / \partial x^2 , \quad (4)$$

which, depending on the expression for $\partial\beta/\partial x$, is either in the form of a simplified Fokker-Planck equation (if $\partial\beta/\partial x$ is a constant) or the Burgers equation if $\partial\beta/\partial x$ depends linearly on C_l . A more rigorous derivation of Eq. (4), with a more detailed expression for the term β , can be found in Zik *et al.* [10]. Equation (4) has also been found to describe automobile traffic flow, which is modeled as a non-Newtonian fluid. In this case the conditions for which $D_{\text{eff}} < 0$ lead to traffic jams and instabilities [13].

In order to account for the reversal in axial segregation with rotation speed illustrated in Fig. 1 the difference between ϕ of the mixed phase and ϕ of the large bead phase must decrease as ω is lowered so that $D_{\text{eff}} > 0$. Measurements of the dynamic angle of repose for homogeneous phases of large beads and for a 50-50 mixture as a function of rotation speed are shown in Fig. 3. The dynamic angle ϕ is determined using photographs of the end plate during rotation and measuring the angle that the freely flowing surface makes with various fixed horizontal markers. Measurements of ϕ for the homogeneous phase of small beads displayed a greater sensitivity to variations of humidity, therefore only the data for the large beads are included in Fig. 3. In addition to the error associated with measuring a moving surface, the avalanching surface does not form a perfect plane, due to centrifugal forces and wall friction which give the flowing surface an s-shaped contour at high rotation speeds [14]. Consequently the values of ϕ in Fig. 3 are the averaged values of ~ 10 -20 separate angular measurements for each phase

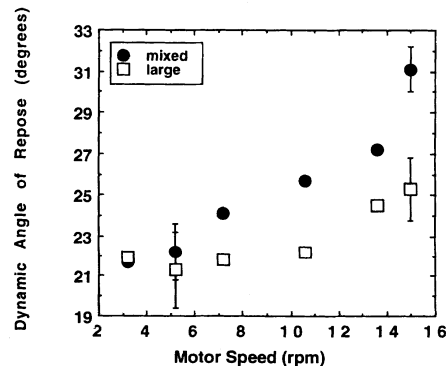


FIG. 3. Plot of the dynamic angle of repose for the mixed phase and the large glass beads against horizontal rotation speed.

at each ω [15]; the standard deviations are indicated for $\omega = 5$ and 15 rpm. Figure 3 shows that the average value of ϕ for the mixed phase is clearly larger than for the segregated phase when $\omega = 15$ rpm, and is nearly equal to the large bead ϕ value at 5 rpm. Since β is a monotonic function of the difference between ϕ of the mixed and segregated phases, we expect $\beta > D$ at 15 rpm, which leads to $D_{\text{eff}} = D - \beta < 0$ and axial segregation while decreasing the rotation speed to 5 rpm, where $\beta = 0$ and $D_{\text{eff}} = D > 0$. A positive D_{eff} will cause the segregated phases to mix together, while if we start with a mixed phase at $\omega = 5$ rpm we do not observe segregation regardless of the rotation time. Moreover, measurements of the dynamic angle of repose for homogeneous phases and a 50-50 mixture of "medium" size glass beads (diameter ~ 1.5 -2 mm) and sand (diameter ~ 250 -500 μm) which exhibits a nonreversible axial segregation find that the ϕ of the mixed phase is larger than ϕ for the glass beads at all rotation speeds. The reversible axial segregation is therefore a direct result of the variation of $\Delta\phi$ with ω , and in particular that $\Delta\phi = 0$ for small but finite rotation speeds.

The dynamic angle of repose for a granular material

depends on, among other factors, the rotation speed, the particle's size, mass, and roughness, the kinetic friction between particles and between the granular media and the wall of the rotation drum, as well as the heterogeneity of the material [4,7,14]. Monodisperse glass beads contain a higher void fraction than a mixture of beads of different sizes; the latter will be better able to sustain a shear flow and consequently will have a larger dynamic angle of repose [6]. Further studies of rotating granular mixtures as a function of relative concentrations and changing initial and boundary conditions should improve our understanding of the factors governing both the dynamic angle of repose and the axial segregation phenomenon.

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- [1] H. M. Jaeger and Sidney R. Nagel, *Science* **255**, 1523 (1992).
 - [2] James B. Knight, H. M. Jaeger, and Sidney R. Nagel, *Phys. Rev. Lett.* **70**, 3728 (1993).
 - [3] Y. Oyama, *Bull. Inst. Phys. Chem. Res. (Tokyo)*, **Rep. 5**, 600 (1939).
 - [4] M. B. Donald and B. Roseman, *British Chem. Eng.* **7**, 749 (1962).
 - [5] J. Bridgwater, N. W. Sharpe, and D. C. Stocker, *Trans. Inst. Chem. Eng.* **47**, T114 (1969).
 - [6] J. Bridgwater, *Powder Tech.* **15**, 215 (1976).
 - [7] S. Das Gupta, D. V. Khakhar, and S. K. Bhatia, *Powder Tech.* **67**, 145 (1991); *Chemical Eng. Sci.* **46**, 1513 (1991).
 - [8] S. Fauve, C. Laroche, and S. Douady, in *Physics of Granular Media*, edited by Daniel Bideau and John Dodds (Nova Science, Commack, NY, 1991), p. 277.
 - [9] Stuart B. Savage, in *Disorder and Granular Media*, edited by D. Bideau and A. Hansen (North-Holland, Amsterdam, 1993), p. 255.
 - [10] O. Zik, Dov Levine, S. G. Lipson, S. Shtrikman, and J. Stavans (unpublished).
 - [11] S. F. Edwards and Anita Mehta, *J. Phys. (Paris)* **50**, 2489 (1989); Anita Mehta and S. F. Edwards, *Physica A* **157**, 1091 (1989); **168**, 714 (1990).
 - [12] K. M. Hill and J. Kakalios (unpublished).
 - [13] M. J. Lighthill and G. B. Whitham, *Proc. R. Soc. London Ser. A* **229**, 317 (1955); Paul I. Richards, *Oper. Res.* **4**, 42 (1956); Robert E. Chandler, Robert Herman, and Elliott W. Montroll, *Oper. Res.* **6** 165 (1958); G. F. Newell, *ibid.* **9**, 209 (1961).
 - [14] Jean Rajchenbach, *Phys. Rev. Lett.* **65**, 2221 (1990).
 - [15] The mixed phase was remixed by hand at the higher rotation speeds every few minutes to avoid segregation effects.

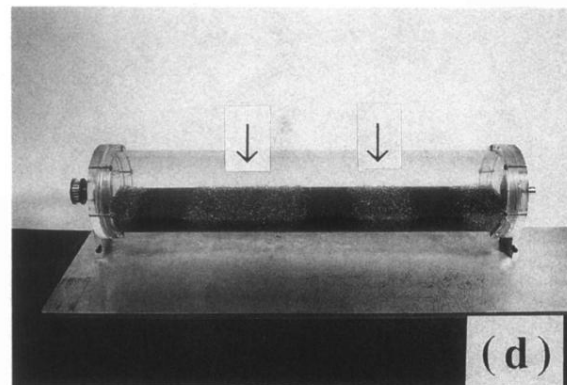
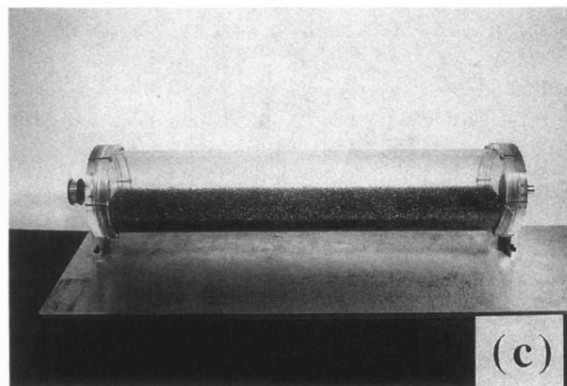
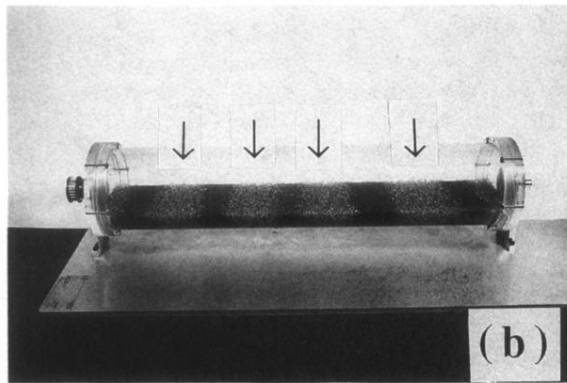
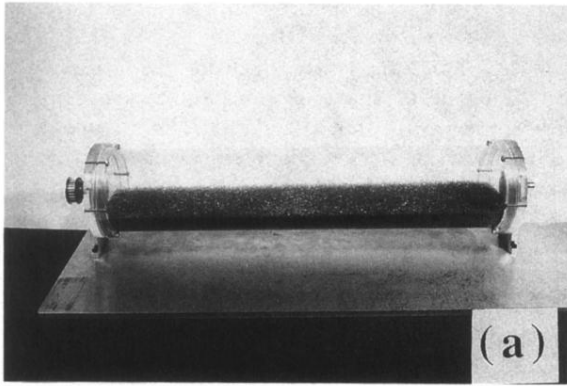


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