Dynamics of axial separation in long rotating drums

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We propose a continuum description for the axial separation of granular materials in a long rotating drum. The model, operating with two local variables, concentration difference and the dynamic angle of repose, describes both initial transient traveling wave dynamics and long-term segregation of the binary mixture. Segregation proceeds through ultra-slow logarithmic coarsening.

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The collective dynamics of granular materials recently have been attracting much interest [1,2]. Intrinsic dissipative nature of interaction among macroscopic particles sets granular matter apart from familiar gaseous, liquid, or solid states. One of the most fascinating features of heterogeneous granular materials is their ability to segregate under external agitation instead of mixing as would be expected from thermodynamics. Essentially any variation in mechanical properties of particles (size, shape, density, surface roughness) may lead to their segregation. Long rotating drums partially filled with a mixture of grain sizes exhibit both radial and axial size segregation [3,5,6]. In radial segregation grains of one type (for grains of different sizes, the smaller ones) build up a core near the axis of rotation. Axial segregation often follows radial segregation, with the mixture of grains separating into bands according to size arranged along the axis of the drum. Axial segregation leads to either a stable array of concentration bands, or to the complete segregation [7].

Granular flow in a rotating drum is different from conventional fluid motion. In the bulk, particles perform solid body rotation around the drum axis until they reach the free surface. Then they slide down within a thin near-surface layer [3,8]. The surface has a $S$-curved shape and its average slope is determined by the dynamic angle of repose. Since there is almost no inter-particle motion in the bulk, the segregation predominantly occurs within the fluidized near-surface layer. The radial segregation occurs during first few revolution of the drum. For long drums (length much larger than radius), along with radial segregation, axial segregation occurs at much later stages (after hundreds of revolutions). Recent experiments [3,5,8] revealed interesting features of axial segregation. At early stages, small-scale perturbations travel across the drum in both directions (it was obvious from the dynamics of pre-segregated mixtures [8]), while at later times more long-scale static perturbations take over and lead to quasi-stationary bands of separated mixture. Bands of segregated materials interact at a very long time scale and exhibit logarithmic coarsening [8].

This process can be accelerated in a drum of a helicoidal shape. The bands can be locked in a drum with the radius modulated along the axis [8].

Most of the theoretical models of segregation agree in that the underlying reason for segregation is the sensitive dependence of the surface slope or shape on the relative concentration of different particles in the mixture [3]. In Ref. [3] a simple theory was proposed of segregation in thin surface flow driven by the local slope, combined with conservation of particles. For monodisperse material, the model recovers the $S$-shape of the free surface. For a binary mixture the model yields a nonlinear diffusion equation for the relative concentration of the ingredients along horizontal axis. Axial segregation occurs when the diffusion coefficient turns negative. This model yields a significant insight into the nature of the instability leading to the segregation, however since it is based on a first-order diffusion equation, it fails to describe the traveling waves observed at the early stages of axial segregation [8].

In this Letter we propose a continuum model which describes consistently the early phase of segregation with traveling waves as well as the later stage of segregation characterized by slow merging of bands of different particles. Our model predicts slow (logarithmic) coarsening of the segregated state. The dynamics of segregation shows striking similarity with the experiments of Ref. [3].

Let us consider a mixture of two sorts of particles, $A$ and $B$, of which $A(B)$ corresponds to particles with larger (smaller) static repose angle. Our model operates with two continuum variables: the relative concentration of particles $c = (c_A - c_B)/(c)$, and local dynamic repose angle $\theta$. Here $c_{A,B}$ are local partial concentrations of particles, and $\langle c \rangle = \langle c_A + c_B \rangle$ is an average over whole system total concentration. We assume that $c$ and $\theta$ are functions of longitudinal coordinate $x$ and time $t$. In fact, concentration also depends on the radial coordinate, however in this study we are concerned with axial segregation, and will operate with quantities integrated over cross-sections of the long drum.
The first equation represents conservation of the relative concentration $c$ in the binary mixture:

$$\partial_t c = -\partial_x(-D\partial_x c + g(c)\partial_x \theta) \quad (1)$$

The first term describes diffusion flux (mixing), and the second term describes differential flux of particles due to the gradient of dynamic repose angle. Similar term with specific function $g(c) = G_0(1 - c^2)$ (in our notation) was first derived in Ref. 6 from the condition of detailed flux balance. The proportionality constant $G_0$ depends on the physical properties of the grains (see Ref. 8). For simplicity, the constant $G_0$ can be eliminated by the scaling of distance $x \to x/\sqrt{G_0}$. The sign $+$ before this term means that the particles with the larger static repose angle are driven towards greater dynamic repose angle. As will be shown below, this differential flux gives rise to the segregation instability. Since this segregation flux vanishes with $g(c) |c| \to 1$ (which correspond to pure $A$ or $B$ states), it provides a natural saturation mechanism for the segregation instability.

The second equation describes the dynamics of $\theta$

$$\partial_t \theta = \alpha(\Omega - \theta + f(c)) + D_\theta \partial_{xx} \theta + \gamma \partial_{xx} c \quad (2)$$

Here $\Omega$ is the angular velocity of the drum rotation, and $f(c)$ is the static angle of repose which depends on the relative concentration. According to our definition of $c, f(c)$ is an increasing function of relative concentration [8]. Since the angle of repose as a function of the concentration $c$ changes typically in a small range, we can approximate the function $f(c)$ by linear dependence $f(c) = F + f_0 c$. The constant $F$ can be eliminated by the substitution $\theta = \theta - F$. First term in the r.h.s. of Eq. (2) describes the local dynamics of the repose angle ($\Omega$ increases the angle, and $-\theta + f(c)$ describes the equilibrating effect of the surface flow), and the term $D_\theta \partial_{xx} \theta$ describes axial diffusive relaxation. The last term, $\gamma \partial_{xx} c$, represents the lowest-order non-local contribution from the inhomogeneous distribution of $c$ (the first derivative $\partial_x c$ cannot be present due to reflection symmetry $x \to -x$). As we will see later, this term gives rise to the transient oscillatory dynamics of the binary mixture.

The stationary uniform state of the system is $c = c_0; \theta_0 = \Omega + f_0 c_0$ where $c_0$ is determined by initial conditions. $\theta_0$ has a meaning of the stationary dynamic repose angle, which in the limit $\Omega \to 0$ approaches the static repose angle [8].

Let us consider the stability of the uniform state,

$$c = c_0 + C e^{\lambda t + ikx}$$

$$\theta = \theta_0 + \Phi e^{\lambda t + ikx} \quad (3)$$

To linearize the system, we need to expand the function $g(c)$ near $c_0$. The stability properties depend on the values of $f_0$ and $g_0 \equiv g(c_0)$. The eigenvalues $\lambda_{1,2}$ are found from the following equation,

$$(\lambda + Dk^2)(\lambda + D\theta k^2 + \alpha) - g_0(\alpha f_0 k^2 - \gamma k^4) = 0 \quad (4)$$

At $k \to 0$ one finds $\lambda_1 = -\alpha - (D_\theta + g_0 f_0)k^2$; $\lambda_2 = (g_0 f_0 - D)k^2$. Asymptotic expansion at $k \to \infty$ yields

$$\lambda_{1,2} = \frac{1}{2} \left(-D_\theta - D \pm \sqrt{(D_\theta - D)^2 - 4g_0\gamma}\right)k^2. \quad (5)$$

It is easy to see that if $g_0 f_0 > \alpha D$, long-wave perturbations are unstable ($\lambda_2 > 0$), and if $g_0 \gamma > (D_\theta - D)^2/4$, short-wave perturbations oscillate and decay (eigenvalues $\lambda_{1,2}$ are complex conjugate with negative real part). The full dispersion curve $\lambda(k^2)$ for particular values of parameters is plotted in Fig. 1. This curve is consistent with the measurements in Ref. 6. Indeed, at small wavelengths the perturbations travel, and their phase velocity $v_{ph} = \text{Im}\lambda/k$ grows linearly with $k$. At small $k$, frequency and phase velocity turn into zero and remain zero for all long-wave perturbations $k < k_s$. Perturbations in the range $0 < k < k_s$ where $k_s = (\alpha (g_0 f_0 - D)/(g_0 \gamma + DD_\theta))^{1/2} < k_s$, grow exponentially and there is an optimal wavenumber of the fastest growing perturbations $k_0$. Oscillating perturbations with $k > k_s$ always decay, which seems to be in agreement with experimental data, although direct experimental measurement of Re$\lambda(k)$ is lacking.

We performed numerical simulations of Eqs. (1,2) using pseudo-spectral split-step method with periodic boundary conditions. We used up to 512 mesh points in our numerical procedure. The following form of nonlinear functions in Eqs. (1,2) was implemented: $g(c) = 1 - c^2$ and $f(c) = f_0 c \Omega$ [10]. The dynamics of the initially pre-separated state with wavenumber $k > k_s$ in a system with size $L = 60$ is shown in Fig. 2. In agreement with experiments [6], short-wave initial perturbation produce decaying standing waves, which later are replaced by quasi-stationary bands (Fig. 2a). The bands are separated by sharp interfaces which are very weakly attracted to each other. In fact, in simulations with parameters corresponding to Fig. 2a we were not able to detect interface merging at all in a reasonable simulation time. However at higher rates of diffusion and dissipation, corresponding to different experimental conditions (rotation frequency, filling factor, etc), the interaction becomes more significant, and it leads to band merging and overall pattern coarsening (see Fig. 2b). In Fig. 2b we present a number of bands as a function of time for this run.

Fronts separating bands of different grains, can be found as stationary solutions of Eqs. (1,2). In an infinite system one finds from stationary Eq. (1) $\theta = \theta_0 + DG(c)$, where $G(c) = \int [g(c)]^{-1} dc = -\frac{1}{2} \log \frac{1}{1+c}$ and $\theta_0$ is an integration constant. Plugging this expression in Eq. (2), we obtain the second order differential equation for $c$ (for a symmetric solution one chooses $\theta_0 = \Omega$),

$$\frac{d}{dx} \left[ \left( \gamma + \frac{DD_\theta}{1-c^2} \right) \frac{dc}{dx} \right] + \alpha f_0 c + \frac{\alpha D}{2} \log \frac{1-c}{1+c} = 0. \quad (6)$$
It is easy to see that this equation possesses an interface solution. The asymptotic behavior of this solution can be found in the limit $D \ll f_0$, when the states on both sides of the interface are well segregated ($|c(x \to \pm \infty)| \to \pm 1$). In this limit, far away from the interface $1 - |c| \propto \exp(-x/d_0)$ where $d_0 = \sqrt{\frac{2}{D}} + \frac{D}{d} \exp(-\frac{D}{2d})$. As seen from this formula, the characteristic front width vanishes as $D, D_0 \to 0$. This result could be anticipated, as in the absence of diffusion nonlinearity $g(c)$ drives the system towards complete segregation.

Slow logarithmic coarsening of the segregated state can be understood in terms of the weak interaction of fronts. Since the asymptotic field of the front approaches equilibrium value of the concentration exponentially fast, we expect in general exponentially weak interaction between the neighboring fronts leading to logarithmic times for the front annihilation.

The problem of front interaction has been recently considered \cite{12} for the Cahn-Hilliard model which arises in relaxation phase ordering kinetics \cite{11}. As in the Cahn-Hilliard model, the order parameter (here concentration $c$) is a conserved quantity, therefore front interaction must conform to a global constraint. In fact, in a particular case $g(c) = 1$, $D = 0$, $D_0 = 0$ our model \cite{1,2} can be reduced to a single equation for the relative concentration

$$\partial_t c + \partial_x c = -\partial_{xx}(f(c) + \partial_{xx}c)$$

which differs from the Cahn-Hilliard model only by the first term in the l.h.s. This term describes non-potential effects, including traveling waves discussed above. For the slow process of the band relaxation in the long-time limit this term can be dropped. Front solutions within the Cahn-Hilliard equation exist for $N$-shaped functions $f(c)$, for example, a cubic polynomial $f(c) = 2(c - c^3)$. In this case stationary interface solution is found analytically as $c(x) = c_0 + \tanh(x - x_0)$, and interaction of the interfaces can be analyzed. This analysis \cite{12}, predicts that a single band of positive $c$ in the sea of negative $c$ (two fronts) can either annihilate or reach a stationary width depending on the initial distance between fronts.

If a number of fronts is greater than 2, front interaction leads to their annihilation and pattern coarsening. The interaction between fronts due to their tail overlapping is exponentially weak. The rate of front interaction can be determined using multiple scale analysis. In the simplest case of three equidistant fronts, two outer fronts collide with the central front and disappear after time $T \propto d \exp(d/d_0)$ where $d$ is the initial distance between fronts. Thus, for multi-band structure, the number of fronts $N$ (proportional to the inverse average distance between fronts) decreases approximately as a logarithmic function of time ($N = 1/d \sim 1/(const + d_0 \log T)$). Since interfaces in our more complicated model have similar exponentially small tails, we anticipate a similar logarithmic law of band coarsening. This dependence indeed agrees with our numerical simulations (see Fig. 3).

In conclusion, we proposed a simple continuum model for axial segregation of binary granular mixtures in long rotating drums. The model operates with two local dynamical variables, relative concentration of two components and dynamic repose angle. The dynamics of our model shows qualitative similarity with the experimental observations of initial transients and long-term segregation dynamics \cite{1,2}. It captures both initial transient traveling waves and subsequent onset band structure. The dispersion relation for slightly perturbed uniform state \cite{4} qualitatively agrees well with observations \cite{4}, and can serve for fitting the model parameters. The logarithmic coarsening of the quasi-static band structure which follows from our model, deserves experimental verification. This coarsening is typical for systems with exponentially weak attractive interaction among defects or interfaces, as in the phase ordering kinetics described by the Cahn-Hilliard model. Our simulations also showed that the model qualitatively reproduces more complicated phenomenology of the separation process reported in Ref. \cite{4}. In particular, periodic modulation of the drum radius, modeled in our approach by periodic variation of $\Omega$, leads to band locking. Breaking of the $x \to -x$ symmetry by term $\partial_\theta \epsilon$, introduced in r.h.s. of Eq. \cite{4}, results in complete segregation, similarly to the dynamics of grains in the drum with helicoidal shape.

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[9] In fact, our model is applicable only for intermediate values of $\Omega$, since for very small $\Omega$, the regime of continuous surface flow is replaced by avalanches, and at large $\Omega$, the flow of particles in not confined to the narrow near-surface region.

[10] Nonlinearity in function $f(c)$ in principle can also contribute to the saturation of the segregation instability. However, since measurements [8] show little nonlinearity of $f(c)$, the simplest form of $f(c)$ is sufficient. Simulations with nonlinear function $f(c) = f_0 \tanh c$ did not reveal any quantitative difference with respect to linear approximation.

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**FIG. 1.** Dispersion relation $\lambda(k)$ for the small perturbation of the uniform state $c_0 = 0$ at $f_0 = 40$, $D = 0.05, D_\theta = 0.1, \alpha = 0.025, \gamma = 1$. Perturbations are unstable at $k < k_c = 0.994$ and oscillate (and decay) at $k > k_c = 1$.

**FIG. 2.** Space-time diagrams of the evolution of the pre-separated state ($c(x,0) = c_0 + c_i \cos(k_0x)$) with $c_0 = 0$, $c_i = 0.95$, and $k_0 = 1.79 > k_*$: a) initial transient, At times $t < 15$ the initial perturbations excite decaying standing wave (superposition of left- and right-traveling waves), and at larger times $t > 15$, aperiodic segregated bands emerge. Parameters of the model are: $D = 0.05, \gamma = 1, \alpha = 0.025, f_0 = 40, L = 60$ and $D_\theta = 0.1$. b) Space-time diagram for long-time evolution, band merging and coarsening during long-time evolution ($0 < t < 10000$) at higher diffusion constants, parameters $D = 0.8, D_\theta = 0.5, \gamma = 1, \alpha = 0.5, f_0 = 2, L = 140$.

**FIG. 3.** Number of fronts $N$ as a function of time (diamonds) and its fit by a function $N = 70/(\ln t - 2.5)$ (long-dashed line). Parameters correspond to Fig.2b.