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Segregation of granular mixtures in the presence of compaction

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Abstract. – We study the segregation phenomena in granular media in the framework of *Tetris*-like models for granular compaction. The models, which focus on the geometrical properties of mixtures, exhibit segregation, *i.e.* separation of particles with different degrees of geometrical frustration. This effect is related to the effective average mobilities of the different particles. We study these effects in the context of an analytical mean-field–like model.

Segregation phenomena in granular media are examples of common and simple non-equilibrium dynamical processes. They occur for instance in granular mixtures, where, under shaking, larger particles rise to the top even if there is no density difference with the other components [1]. As shown in recent experiments [2, 3] and computer simulations [4], size segregation is linked to convective flow inside shaken grains containers. However, there are situations, when convective rolls are absent or less important, in which the mechanical motor underlying segregation effects is more subtle and stochastic in nature [1, 3]: during shaking small particles may occasionally fill the voids generated below the larger particles producing the net effect of their upward motion. This experimental picture is well described by several computer simulations [5-7]. Segregation is also very easily observed in many other apparently heterogeneous situations (see references in [1,8]).

In this paper we describe the mechanisms underlying segregation in the framework of the *Tetris*-like models already introduced to explain density compaction [9] in granular media. The model, defined on a lattice, captures the original idea of the computer game Tetris of difficult parking problems in the packing of objects of different shapes. It consists of a

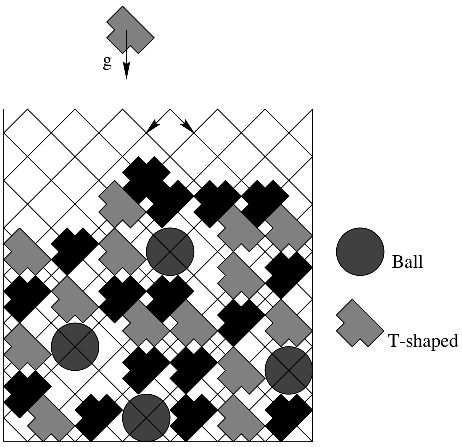

Fig. 1. – A picture of the lattice model considered in the present paper showing the shapes of its particles.

system of particles with different shapes which cannot overlap during their diffusion. In a very general way we can define the model with a complex interaction matrix which introduces, for each particle, a "geometrical frustration", *i.e.* the constraints the particle has to fulfill in the combinations with the neighbor particles.

When subject to gravity and vibrations the model exhibits a very slow relaxation dynamics due to high entropic barriers (originated for geometrical reasons) to be passed by particles. In polydisperse mixtures, *i.e.* in the presence of particles with very different packing properties, the model exhibits segregation, that is a tendency to separate particles with different degrees of frustration. This mechanism underlies a size segregation which is always in competition with other factors like gravity, friction etc. As a consequence of this competition there exists, in general, a complex phase diagram with regions which show different behaviors and drive segregation in different directions.

For the sake of simplicity, but without loss of generality, we consider a model with just three possible shapes for the particles which occupy the sites of a square lattice tilted by 45° (see fig. 1): a spherical one, which prevents any other particle from occupying its nearest neighbors, and two "T"-shaped types of grains which can find arrangements in which they are first neighbors provided they do not violate the constraints of non-overlapping [10]. In this way the "T"-shaped particles are less frustrated than the spherical ones because they impose constraints only on three nearest neighbors. From this point of view it is possible to say that spherical particles are bigger than the "T"-shaped ones. We will denote with c the fraction of spherical particles with respect to the total number of particles. Moreover, we consider periodic boundary conditions in the horizontal direction (cylindrical geometry) and a rigid plane at its bottom. The "T"-shaped particles can rotate provided that at least three of their nearest neighbors are empty. This condition is such that for sufficiently high densities the rotation events become negligible and the particles definitely keep their orientation.

We study such a system in the presence of "external vibrations" and "gravity". So we define a dynamics based on the random diffusion of particles in such a way as to preserve the above geometrical constraint of non-overlapping. The particles attempt a move upward (downward) with probability p_{up} (p_{down}) ($p_{\text{down}} + p_{\text{up}} = 1$). The parameter which controls the dynamics is then the ratio $x = p_{\text{up}}/p_{\text{down}}$.

The system is prepared by randomly pouring particles into the box from its top and then

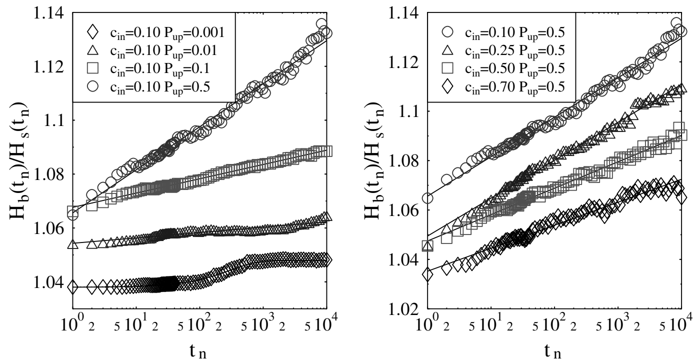

Fig. 2. – The ratio $H_b(t_n)/H_s(t_n)$ grows with the tapping number t_n, showing a net effect of segregation of the big spherical particles on the top of the mixture. *Left*: H_b/H_s for tapping sequences of samples with a fixed relative concentration of the two species of the mixture, $c = 0.1$, shaken at $p_{\mathrm{up}} = 0.001, 0.01, 0.1, 0.5$ (from bottom to top). *Right*: H_b/H_s for sequences of samples shaken at a $p_{\mathrm{up}} = 0.5$ but with different $c = 0.1, 0.25, 0.5, 0.7$ (from top to bottom). The tap duration is $\tau = 10$. The ratio $H_b(t_n)/H_s(t_n)$ increases faster for larger p_{up} and for lower c.

letting them fall down, one at a time, performing an oriented random walk ($p_{\mathrm{up}} = 0$). The system is then subjected to a vibration procedure realized as a two-step dynamical Monte Carlo sequence which mimics the experimental procedure of tapping. In the first step one fixes $x \neq 0$ for $t \in [0, \tau]$ (we fix $\tau = 10$, in units of moves attempted per particle). In the second step we fix $x = 0$ and we let the system attain a *static* configuration, *i.e.* a configuration in which particles cannot move anymore. This procedure is repeated for each single tap. The measured quantities are averaged over 50 different random realizations for a square lattice of size 30×30. We checked that our results are strongly robust with respect to lattice size effects and with respect to variations in the Monte Carlo procedure. In particular one gets the same results with a procedure of continuous vibration in which $x = \mathrm{const} \neq 0$.

As stated above, the tapping sequence causes the compaction of the system. The static bulk density $\rho(t_n)$ measured after each tap ($t_n = n$ is the time corresponding to the n-th tap, measured in number of taps) of each single species in the lower 25% of the box (ρ_s for the small "T"-shaped and ρ_b for the spherical particles) increases with t_n according to the inverse logarithmic law experimentally found in [11] (see also ref. [9, 11-14]). Also the average heights of different species $H(t_n)$ as a function of time ($H_s(t_n)$ for the "T"-shaped and $H_b(t_n)$ for the spherical particles), consistently with the observations about compaction, decrease with t_n and they are very well fitted by the inverse logarithmic law quoted above [11]:

$$H(t_n) = H_\infty + \frac{\Delta H_\infty}{1 + B \log(t_n/\tau_0 + 1)} . \tag{1}$$

For the tapping sequence previously described, with $c = 0.25$ and $p_{\mathrm{up}} = 0.5$, the values of the fit parameters of eq. (1) for the average heights are $H_\infty = 11.1$, $\Delta H_\infty = 4.3$, $B = 0.23$ and $\tau_0 = 0.09$ for $H_s(t_n)$, and $H_\infty = 13.3$, $\Delta H_\infty = 2.0$, $B = 0.67$ and $\tau_0 = 0.55$ for $H_b(t_n)$.

These parameters show that the compaction of the two types of particles is different. In particular the ratio $H_b(t_n)/H_s(t_n)$ grows with t_n (fig. 2). We recorded two types of tapping sequences: one with a given relative concentration of the species $c = 0.1$ shaken with different values of p_{up}, and the other performed with a fixed value of $p_{\mathrm{up}} = 0.5$ and different values of c. The tap duration is always $\tau = 10$ in both cases. The ratio $H_b(t_n)/H_s(t_n)$ increases faster the larger p_{up} is and the lower c is. These results exhibit a net effect of segregation of the big

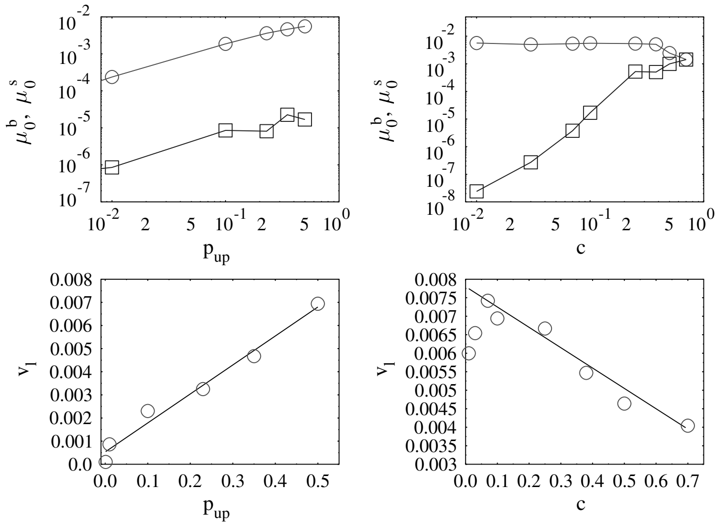

Fig. 3. – *Top left and top right*: values of the average mobilities of the two kinds of particles μ_0^s (circle), μ_0^b (square) in the two sequences at fixed $p_{\rm up}$ or fixed c. *Bottom left and bottom right*: the segregation "velocity", $v_l(p_{\rm up}, c)$, as a function of the vibration amplitude $p_{\rm up}$ and of the relative species concentration c corresponding for two sequences at fixed $p_{\rm up}$ or fixed c.

spherical particles on the top of the mixture.

Next we study the average particle *mobility* $\mu(h, t_n)$ defined as the average number of successful moves per particle at depth h divided by the total number of attempted moves during the n-th tap. In our system $\mu(h, t_n)$ strongly depends on the depth (measured in lattice units). To deal with quantities with reasonable statistics, we introduce the quantities $\mu_0(t_n)$, defined as the average mobilities over the lower 33%. Figure 3 shows the average mobilities $\mu_0^s(t_n = 10^4)$ and $\mu_0^b(t_n = 10^4)$ in the two quoted sets of sequences at fixed $p_{\rm up}$ (top-left) and fixed c (top-right). From our data it is evident that the big spherical particles, which segregate on the top of the system, have a lower mobility with respect to the others. More in general we expect that the mobility associated to each particle is smaller the bigger its geometrical frustration is. A rough estimate of the velocity of segregation in our model can be obtained by looking at the ratio $H_b(t_n)/H_s(t_n)$ which can be fitted with the function $H_b(t_n)/H_s(t_n) = h_0 + v_l \log(t_n)$. Figure 3 reports the values of the "velocity" of segregation, $v_l(p_{\rm up}, c)$, *i.e.* the slope of fig. 2, as a function of $p_{\rm up}$ or c for the two sequences at fixed $p_{\rm up}$ (bottom left) or fixed c (bottom right). This clarifies the qualitative observation that the segregation is faster the higher $p_{\rm up}$ is and the lower c is. The different behaviors with $p_{\rm up}$ and c can be linked to the differences of the mobility of the two kinds of particles during the corresponding tapping sequence. In particular, in fig. 3 for the set of sequences with fixed $c = 0.1$ we see that the increase of v_l with $p_{\rm up}$ corresponds to an increase of the mobility of both kinds of particles even though their ratio remains almost constant (top left). On the other hand, in the set of sequences with fixed $p_{\rm up} = 0.5$, the decrease of v_l with c corresponds to an increase of the ratio μ_0^b/μ_0^s. The velocity of segregation is thus linked to both the values of global mobility and the relative mobility of the different types of particles. When the mobilities of the two species are very close, the segregation is highly suppressed (top right). In this way we have a very simple picture of the effect on segregation of the different mobilities of the mixture components.

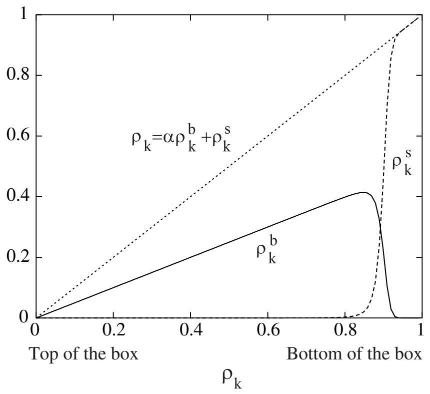

Fig. 4. – Asymptotic density profile for the two species of particles in the N-planes model described in the text. The effect of segregation of the more frustrated particles (label b) on top of the less frustrated ones (label a) is clearly visible.

The results obtained here about segregation can be further clarified within the context of a simple mean-field–like model introduced in [9]. It consists of a system of N-planes which can exchange particles with difficult parking problems. The model, which ignores correlations among particles rearrangements and the problems related to the mechanical stability of the assembly, just takes into account the geometrical frustration, say the difficulty for the particles to find a large enough space to diffuse. In the case of a polydisperse medium with two species of grains, the master equations for densities ρ_k^b and ρ_k^s on a generic plane k, except for the plane $k=0$, are given by

$$\partial_t \rho_k^{b,s} = M^{b,s}(\rho_k)[\rho_{k-1}^{b,s} p_{\rm up} + \rho_{k+1}^{b,s} p_{\rm down})] - \rho_k^{b,s}[M^{b,s}(\rho_{k-1}) p_{\rm down} + M^{b,s}(\rho_{k+1}) p_{\rm up}] \,, \qquad (2)$$

where $\rho_k = \rho_k^s + \alpha\rho_k^b$ is the total density on the k-th plane and $M^{b,s}(\rho_k)$ are the probabilities for a particle s or b respectively, landing on the plane k, to fit the local geometrical environment. In this sense $M^{b,s}(\rho)$ are the analog of the mobility μ we have introduced above and they take into account the cooperative effects on the dynamics generated by the frustration. A possible functional form for $M^{b,s}(\rho)$ (see for a similar approach [9,13,14]), is given by $M^s(\rho) \sim (1-\rho)\exp[-\rho/(1-\rho)]$ and $M^b(\rho) \sim (1-\rho)^\alpha \exp[-\alpha\rho/(1-\rho)]$ with $\alpha \geq 1$. The parameter α quantifies how much the shape and the dimensions of the particle frustrate its motion. The higher α is the higher the geometrical frustration felt by the corresponding particle will be. For instance in the problems of parking of r-mers (*i.e.* segments of length r) on a line, the probability for each r-mer to find a sufficient space to land is $\exp[-r\rho/1-\rho]$, where ρ is the occupation density on the line [13]. α plays thus the role of the linear size of the corresponding particle. Let us stress how this mean-field model evolves to an asymptotic stationary state with a shaking procedure which is in the same spirit of the continuous vibration in the *Tetris*-like model presented above. The asymptotic stationary solution of the system (2) can be obtained exactly by putting $\partial_t \rho_k^{b,s} = 0$. In this way one gets, for the asymptotic density profile of the two species:

$$\rho_k^b = \rho_k \frac{h_0(1-\rho_k)^{\alpha-1} e^{-\frac{\alpha-1}{1-\rho_k}}}{1+\alpha h_0 (1-\rho_k)^{\alpha-1} e^{-\frac{\alpha-1}{1-\rho_k}}} \rho_k^s = \rho_k - \alpha\rho_k^b \,, \qquad (3)$$

where $\rho_k = \rho_k^s + \alpha\rho_k^b$, $h_0 = h_0(\rho_0^b, \rho_0^s)$ is a constant which can be estimated by using the total mass of the system $h_0(\rho_0^b, \rho_0^s) = (1-\rho_0)^{1-\alpha} e^{\frac{\alpha-1}{1-\rho_0} \frac{\rho_0^b}{\rho_0^s}}$. ρ_k is given by a non-trivial function of k that we report in [15]. Here it is enough to say that ρ_k is a monotonically decreasing function with the maximum for $k = 0$. Figure 4 reports an example of the asymptotic stationary solution obtained from (3) with $\alpha = 2$ and $h_0 = 10^5$. In particular it reports ρ_k^b and ρ_k^s as a function of ρ_k. Due to the decreasing character of ρ_k with the height k the right-hand side of the figure, $\rho_k \simeq 1$, corresponds to the bottom of the box, whereas the left-hand side, $\rho_k \simeq 0$, corresponds to the top. In this way the effect of segregation of the "big" particles on the top of the "small" ones is clearly visible. The introduction of two different weights for the two species, say $p_{\rm up}^{b,s}$ and $p_{\rm down}^{b,s}$, gives rise to a rich phase space $(p_{\rm up}^b, p_{\rm up}^s, \alpha)$ with different regions corresponding to different behaviors for segregation [15].

One question one could address is whether there exists a variational principle driving the segregation phenomena. In other words one could ask if, in analogy with what happens for a liquid system, there exists some free-energy-like functional minimized (Lyapunov functional) by the dynamical evolution. In a very general way it is possible to write explicitly a Lyapunov functional for the system of equations (2) (and of course for the monodisperse case), say the functional which decreases monotonically along the trajectories of the motion. This functional can be cast in the form of a free-energy-like function:

$$F = \sum_{k=0}^{\infty} [\gamma(x) k \rho_k - S(\rho_k^b, \rho_k^s)] , \tag{4}$$

where the *entropic* contribution is given by

$$S(\rho_k^b, \rho_k^s) = -\rho_k^s(\log(\rho_k^s) - 1) - \rho_k^b(\log(\rho_k^b) - 1) - (1-\rho_k)(\log(1-\rho_k) - 1) + \log(1-\rho_k) \tag{5}$$

and with $\gamma(x) = \log(1/x)$. Starting from eq. (4) we can find the unique asymptotic stationary solution, say the asymptotic density profile for the two species. The uniqueness of this solution is guaranteed by the convexity of F. Using a standard Lagrange multiplier method one gets the same results shown in (3). Equation (5) makes it evident how in the dynamics of granular media entropic effects are far from being negligible and the global behavior is given by a complex interplay between gravitational and entropic effects [16].

In summary we have studied segregation phenomena in the framework of a recently introduced *Tetris*-like model which takes into account geometrical effects of frustration in the rearrangement processes in granular media. In particular we have related the microscopic motor of segregation to a local mobility of the grains which in its turn is directly related to the geometrical frustration felt by the grains themselves. Furthermore we have shown, in the framework of a mean-field model of N-planes exchanging particles with difficult parking problems, how segregation can be related rigorously to the minimization of a free-energy-like functional. This last point deserves further investigations in order to clarify the thermodynamic properties of granular media.

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