Segregation in fluidized versus tapped packs

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We compare the predictions of two different statistical mechanics approaches, corresponding to different physical measurements, proposed to describe binary granular mixtures subjected to some external driving (continuous shaking or tap dynamics). In particular we analytically solve at a mean field level the partition function of a simple hard sphere lattice model under gravity and we focus on the phenomenon of size segregation. We find that the two approaches lead to similar results and seem to coincide in the limit of very low shaking amplitude. However they give different predictions of the crossovers from Brazil nut effect to reverse Brazil nut effect with respect to the shaking amplitude, which could be detected experimentally.

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Segregation of hard sphere mixtures is a relevant and historically debated problem (see \cite{1} and refs. therein). The phase behavior of these systems is still hotly debated and it is still controversial whether or not hard sphere mixtures segregate in absence of gravity. In the last decade a great attention has been devoted to the study of the problem of vertically shaken granular mixtures under gravity. It was observed that such systems can mix or, under different conditions, segregate their components spontaneously according to criteria which are still largely unclear, although of deep practical and conceptual relevance \cite{5,8,11}. Rosato et al. \cite{5} demonstrated via molecular dynamics simulations that in some cases large spheres segregate to the top and small spheres to the bottom of the container when subjected to shaking. This phenomenon is commonly called Brazil Nut Effect (BNE) (while the opposite one, i.e. large spheres on the bottom and small ones on the top, is known as Reverse Brazil Nut Effect (RBNE) \cite{6,8}). The authors suggested an interpretation of the BNE as a geometric effect (called “percolation”) where small grains pass through the holes created by the larger ones \cite{6,8}. Along with geometry, dynamical effects associated with grains, such as inertia \cite{5} or convection \cite{9}, which bring large particle up but does not allow them to re-enter in downstream, were also shown to play a role \cite{10}.

Recent results have however outlined that segregation processes can involve “global” mechanisms such as “condensation” \cite{6} or phase separation \cite{10,11}. This suggested a change of perspective on the issue and the idea to formulate a statistical mechanics description of these phenomena. In Refs. \cite{6,14} a statistical mechanics approach was proposed to describe segregation in granular mixtures. Assuming that, as in standard thermal systems, the grain kinetic energy may play the role of the bath temperature, $T_{bath}$, the system was approximated as a standard fluidized gas of elastic hard spheres kept at a given temperature, $T_{bath}$, dependent on the shaking amplitude. This approach may be appropriate to describe segregation phenomena in granular mixtures under continuous shaking, but not systems subjected to a “tap dynamics” (where the energy is pumped into the system in pulses, and the measures are performed when the system is at rest). Following the idea, originally suggested by Edwards \cite{15}, and recently further studied \cite{16} (see \cite{3} for a review), it is possible to develop a statistical mechanics description of granular mixtures under such a dynamics. Edwards’ assumption consists in the hypothesis that in a granular system under taps, time averages coincide with suitable ensemble averages over the “mechanically stable” states, i.e., those where the system is found still. Recently, it was shown \cite{17} that this statistical mechanics approach holds in good approximation for a schematic lattice model of a hard sphere binary mixture under gravity. In this case two “configurational” temperatures have to be introduced in order to describe the system’s macroscopic states: These temperatures are just the inverse thermodynamic parameters canonically conjugate to the gravitational energies of the two species, and are not related to the grain kinetic energy which is always zero.

In the present paper we study the relation between these two different approaches in the framework of a simple lattice model for hard sphere binary mixtures under gravity. In particular we analytically solve, at the level of Bethe approximation, the partition function of the system in two different cases (which we will refer to as case I and case II): I) All the configurations are allowed, as in a gas (the system is treated as a standard fluidized hard sphere gas under gravity, and it is not at rest); II) in the framework of Edwards’ theory, where the particles are required to be in a stable configuration (system at rest). We derive the mixing/segregation properties as a function of various parameters such as grain masses, sizes, numbers and others, and finally we compare the results obtained in the two cases. We find that in both
cases the system moves from BNE to RBNE increasing the mass ratio $m_2/m_1$ or decreasing the concentration ratio $N_2/N_1$ of the two species, instead opposite behaviors are found increasing the shaking amplitudes (i.e. the temperatures). In particular in case I the system moves from a BNE state to a RBNE increasing the shaking amplitude, and in case II the opposite one is found. Moreover, for some values of the mass ratio $m_2/m_1$, the RBNE is always found (except at very low temperatures) in the case I, while the BNE is always found in the case II. This scenario could be experimentally checked.

The model we consider [13, 17] is a hard sphere binary mixture made up of two species, 1 (small) and 2 (large) with grain diameters $a_0 = 1$ and $\sqrt{2}a_0$, under gravity on a cubic lattice confined in a rigid box. On each site of the lattice we define an occupancy variable, $n_i^z = 0, 1, 2$, respectively if site $i$ at height $z$ is empty, filled by a small or by a large grain. The Hamiltonian is:

$$H = \mathcal{H}_{HC} + m_1 g H_1 + m_2 g H_2,$$

where $H_1 = \sum_{n,z} z \delta n_i^{z+1}$, $H_2 = \sum_{n,z} z \delta n_i^{z-2}$ are the heights of the two species, and $\mathcal{H}_{HC}$ is the hard core potential, preventing two nearest neighbor sites to be both occupied if at least one contains a large grain.

The partition function of the function for the case of the fluidized hard sphere gas subjected to continuous shaking (case I) is given by:

$$Z_I = \sum_{\{r\}} \exp\left[-\mathcal{H}_{HC}(r) + \beta_{\text{bath}} \left( m_1 g H_1 + m_2 g H_2 \right) \right], \quad (1)$$

where the sum is over all microstates $r$ and $\beta_{\text{bath}}$ is the inverse bath temperature.

As shown in [17] the system subjected to tap dynamics is well described by Edwards’ approach. In this case the weight of a given state $r$ is [17]:

$$\exp\left\{-\mathcal{H}_{HC}(r) - \beta_1 m_1 g H_1(r) - \beta_2 m_2 g H_2(r)\right\} \Pi_r,$$

where $\beta_1$ and $\beta_2$ are the variables conjugate respectively to the gravitational energies of the two species. The operator $\Pi_r$ selects mechanically stable states: $\Pi_r = 1$ if $r$ is “stable”, else $\Pi_r = 0$. We adopt a simple definition of “mechanical stability”: a grain is “stable” if it has a grain underneath. For a given grain configuration, $r = \{n_i\}$, the operator $\Pi_r$ has a tractable expression: $\Pi_r = \lim_{K \to \infty} \exp\left\{-K E_{Edw}\right\}$ where $E_{Edw} = \sum_{n,z} \left[ \delta_{n_i^z} \delta_{\bar{n}_i^{z-1}} - \delta_{\bar{n}_i^{z-2}} + \delta_{n_i^{z-1}} \delta_{\bar{n}_i^{z-2}} \right] (1 - \delta_{n_i^{z-1}})$.

The system partition function in this case is given by:

$$Z_{II} = \sum_{\{r\}} \exp\left[-\mathcal{H}_{HC}(r) + \beta_1 m_1 g H_1 + \beta_2 m_2 g H_2\right] \Pi_r, \quad (2)$$

where the sum is again over all microstates $r$ but, due to the projector, only the mechanically stable ones are taken into account. We found that the configurational temperatures, $T_1^{\text{conf}} = \beta_1^{-1}$ and $T_2^{\text{conf}} = \beta_2^{-1}$, increase as function of the tap amplitude, and tend to coincide in the limit of low tap amplitudes [18]. Thus for simplicity in the following we put $\beta^{\text{conf}}_1 = \beta_1 = \beta_2$.

Since the exact calculation of $Z_I$ and $Z_{II}$ is hardly feasible, we evaluate the partition functions at a mean field level. To this aim we consider a generalization of Bethe-Peierls method for anisotropic systems (due to gravity) already used in previous papers [13, 19], i.e., we solve the partition function of the system on the Bethe lattice shown in Fig. 1 by means of recurrence relations (see also [20]). In particular, we consider a 3D lattice box with $H$ horizontal layers (i.e., $z \in \{1, ..., H\}$) occupied by hard spheres. Each layer is a random graph of given connectivity, $k - 1$ (we take $k = 5$). Each site in layer $z$ is also connected to its homologous site in $z - 1$ and $z + 1$ (the total connectivity is thus $k + 1$). The Hamiltonians are the ones above plus two chemical potential terms which control the two species concentrations. Hard core repulsion prevents two particles on connected sites to overlap.

In the following we give the main ideas of the calculation in the case I, and refer to a longer paper [13] in preparation for the details, and the calculation in the case II (see also [13, 19]). The lattice of Fig. 1 is iterated in three different directions, “side”, “up” and “down”. As consequence three different branches exist, and the corresponding partition functions, $Z_{0,s,u,d}^{(z)}$ and $Z^{(z)}_{n,s,u,d}$ restricted respectively to configurations in which the site $i$ is empty or filled by a particle of species $n = 1, 2$ can be defined. The Bethe-Peierls recursion equations, which allow calculation of the partition function, are more easily written in terms of the local “cavity fields” defined by:

$$e^{h_{n,s}^{(z)}} = \frac{Z_{n,s}^{(z)}}{Z_0^{(z)}}, \quad e^{h_{n,u}^{(z)}} = \frac{Z_{n,u}^{(z)}}{Z_0^{(z)}}$$

The fluid phase corresponds to a solution of Bethe-Peierls equations where local fields in each layer are site independent [21]. Such a solution, characterized by horizontal translational invariance, is given by the fixed points of the following equations:

$$e^{h_{n,s}^{(z)}} = e^{\beta_{\text{bath}}(\mu_n - m_n g z)} \left[ \frac{A_n^{(z)}}{S^{(z)}} \right]^{k-2} \frac{U_{n+1}(z-1)}{P_{n+1}(z-1)},$$

$$e^{h_{n,u}^{(z)}} = e^{\beta_{\text{bath}}(\mu_n - m_n g z)} \left[ \frac{A_n^{(z)}}{S^{(z)}} \right]^{k-1} \frac{U_{n+1}(z)}{P_{n+1}(z)}, \quad (3)$$
\[ e^{f_n^{(z)}} = e^{\beta_{\text{bath}}(\mu_n - m_n g z)} \frac{A_n^{(z)}}{S(z)} \frac{D_n^{(z-1)}}{Q(z-1)}, \]

where \( n = 1, 2; \mu_{1,2} \) are the chemical potentials of the two species and \( m_{1,2} \) are the grain masses; \( A_n^{(z)} = 1 + \delta_n e_i^{(z)} \), \( S(z) = 1 + e_i^{(z)} + e_i^{(z+1)} \), \( U_n^{(z)} = 1 + \delta_n e_i^{(z)} \), \( D_n^{(z)} = 1 + \delta_n e_i^{(z)} \), \( P(z) = 1 + e_i^{(z)} + e_i^{(z+1)} \) and \( Q(z) = 1 + e_i^{(z)} + e_i^{(z+1)} \). From the local field the system free energy can be derived:

\[ F = \sum_{z=0}^{H} \Delta F_n^{(z)} - \frac{(k-1)}{2} \sum_{z=0}^{H} \Delta F_{i,2}^{(z)} - \sum_{z=0}^{H-1} \Delta F_{i,1}^{(z)}, \]

where,

\[ e^{-\Delta F_n^{(z)}} = \left( S(z) \right)^{k-1} P(z+1) Q(z-1) \]

\[ + \sum_{n=1,2} e^{\beta_{\text{bath}}(\mu_n - m_n g z)} \left( A_n^{(z)} \right)^{k-1} U_n^{(z+1)} D_n^{(z-1)}, \]

\[ e^{-\Delta F_{i,1}^{(z)}} = P(z+1) + \sum_{n=1,2} e^{f_n^{(z)}} U_n^{(z+1)}, \]

\[ e^{-\Delta F_{i,2}^{(z)}} = 1 + 2e_i^{(z)} + e_i^{(z+1)} + e_i^{(z+2)}. \]

A similar calculation can be developed in case II. In the following instead of using the chemical potential variables, \( \mu_1 \) and \( \mu_2 \), we will use the conjugate variables, \( N_1 \) and \( N_2 \), the number per unit surface respectively of the small and large grains.

From the free energy \( F \), we calculate the density profiles, \( \sigma_{1,2}(z) \), defined by:

\[ \sigma_n(z) = \frac{1}{N} \left( \frac{2r_n}{a_0} \right)^2 \sum_i n_i(z) \delta_{n_i}, \]

where \( N \) is the site number on each layer, \( n = 1, 2 \) corresponds respectively to small and large grains, and \( r_1 \) and \( r_2 \) are respectively the radius of small grains \( (a_0/2) \) and of large ones \( (\sqrt{2}a_0/2) \). In Fig. 2 we compare \( \sigma_{1,2}(z) \) obtained in the two cases for low and high temperatures. At low temperatures the density profiles are quite similar, as expected since, in the case I, the system explores almost exclusively the mechanically stable states. At high temperatures the density profiles are instead much different.

In Fig. 3 \( \Delta H/H \) as a function of \( N_1 \) (for \( N_2 = 0.4 \)) in the case I (blue continuous line on the top) for \( T_{\text{bath}} = 2 \), and in the case II (red dashed line on the bottom) for \( T_{\text{conf}} = 5 \). The broad horizontal lines identify the “mixing region” where \(-0.1 < \Delta H/H < 0.1\).

In Fig. 3 a diagram of the vertical segregation is given in the plane mass ratio - temperature (respectively \( T_{\text{bath}} \) and \( T_{\text{conf}} \) in the two cases) for fixed values of \( N_1 \) and \( N_2 \). The broad lines correspond to the crossover from BNE to “mixing”, and from “mixing” to RBNE (the “mixing region” is defined as \(-0.1 < \Delta H/H < 0.1\)). The lines on the left are those relative to the case I, which approach asymptotically the vertical line \( m_2/m_1 = 1 \) in the limit \( T_{\text{bath}} \to \infty \), as in a point-like bidisperse gas under gravity. At high temperatures in both cases the system moves from BNE to RBNE with a continuous crossover. Increasing the temperatures the mixing regions broaden since the entropic term in the free energy becomes more important, and the curves in the two cases get apart since the differences of the entropic terms increase. At very low temperatures in both cases the mixing region becomes a line due to the presence of a real phase transition in the corresponding model without gravity. In the diagrams small and large grains tend to demix and due to the gravity one of the two species is energetically favored to stay on the bottom depending on the mass ratio. By further deceans.
and state in the plane mass ratio - temperature (respectively \( T_{\text{bath}} \) and \( T_{\text{conf}} \)) for \( N_1 = 0.8 \) and \( N_2 = 0.4 \). The blue lines on the left are relative to the case I, and the red lines on the right are relative to the case II.

It is evident that at given mass ratio the crossover from BNE to RBNE is obtained by varying the shaking amplitude (i.e. the temperatures) in opposite ways in the two cases: In the case I a crossover from BNE to RBNE is found when \( T_{\text{bath}} \) is increased as experimentally observed in \( \Box \), and in the case II a crossover from RBNE to BNE is found when \( T_{\text{conf}} \) is increased. Moreover, for some values of the mass ratio, the RBNE is always found (except at very low temperatures) in the case I, instead the BNE is always found in the case II. This scenario could be checked via molecular dynamics simulations and experiments of binary granular mixtures: Increasing the shaking amplitude of the continuous external shaking (case I) and of the tap dynamics (case II) in fact corresponds respectively to increasing \( T_{\text{bath}} \) and \( T_{\text{conf}} \).

In conclusion we focus on the problem of the vertical size segregation in binary granular mixtures subjected to external driving. We compare the predictions of two statistical mechanics approaches proposed to describe granular materials, the first more appropriate to deal with fluidized systems subjected to continuous shaking and the second with granular materials subjected to tap dynamics. Applying the two approaches to a simple hard sphere lattice model under gravity we find, in contrast to similar features at low shaking amplitudes, very different behaviors (which could be experimentally checked) as the shaking amplitude is increased. This is due to the prevailing of two different mechanisms giving rise to segregation: for the fluidized granular mixture the segregation is mainly energetic driven, instead for the tapped mixture the segregation is mainly entropic driven. A consequence is for example that the BN region in the second case is larger than in the first one, due to the fact that the stable configurations with small grains on the box bottom are more than those with large grains on the bottom.

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[1] M. Dijkstra, R. van Roij and R. Evans, Phys. Rev. E 59, 5744 (1999), M. Fasolo and P. Sollich, Phys. Rev. Lett. 91, 068301 (2003).
[2] J.M. Ottino and D.V. Khakhar, Ann. Rev. Fluid Mech. 32, 55 (2000). T. Shinbrot and F.J. Muzzio, Physics Today March issue, 25 (2000). J. Bridgewater, Chem. Eng. Sci. 50, 4081 (1995).
[3] "Unifying concepts in granular media and glasses", (Elsevier Amsterdam, in press), Edt.s A. Coniglio, A. Fierro, H.J. Herrmann, M. Nicodemi.
[4] J.E. Mobius, X. Cheng, G.S. Karczmar, S.R. Nagel, H.M. Jaeger, cond-mat/0405328
[5] A. Rosato, K.J. Strandburg, P. Prinz, R.H. Swendsen, Phys. Rev. Lett. 58, 1038 (1987).
[6] D.C. Hong, P.V. Quinn, S. Luding, Phys. Rev. Lett. 86, 3423 (2001).
[7] A. P. J. Breu, H.-M. Enssner, C. A. Krueelle, and I. Rehbberg, Phys. Rev. Lett. 90, 014302 (2003)
[8] J. Bridgewater, Powder Technol. 15, 215 (1976). J.C. Williams, Powder Technol. 15, 245 (1976).
[9] T. Shinbrot and F.J. Muzzio, Phys. Rev. Lett. 81, 4365 (1998).
[10] J.B. Knight, H.M. Jaeger, S.R. Nagel, Phys. Rev. Lett. 70, 3728 (1993).
[11] J.T. Jenkins and D.K. Yoon, Phys. Rev. Lett. 88, 194301 (2002).
[12] P.M. Reis and T. Mullin, Phys. Rev. Lett. 89, 244301 (2002). P. M. Reis, G. Ehrhardt, A. Stephenson and T. Mullin Europhys. Lett. 66, 357 (2004).
[13] M. Tarzia, A. Fierro, M. Nicodemi, M. Pica Ciamarra, and A. Coniglio, cond-mat/0405486
[14] J.A. Both and D.C. Hong, Phys. Rev. Lett. 88, 124301 (2002).
[15] S.F. Edwards and R.B.S. Oakeshott, Physica A 157, 1080 (1989). A. Mehta and S.F. Edwards, Physica A 157, 1091 (1989).
[16] M. Nicodemi, Phys. Rev. Lett. 82, 3734 (1999). A. Barrat, J. Kurchan, V. Loreto and M. Sellitto, Phys. Rev. Lett. 85, 5034 (2000). J.J. Brey, A. Prados, B. Sánchez-Rey, Physica A 275, 310 (2000). A. Coniglio and M. Nicodemi, Physica A 296, 451 (2001). D. S. Dean and A. Lefèvre, Phys. Rev. Lett. 86, 5639 (2001). H. A. Makse and J. Kurchan, Nature 415, 614 (2002). A. Fierro, M. Nicodemi and A. Coniglio, Europhys. Lett. 59, 642 (2002). G. De Smedt, C. Godreche, J.M. Luck, Eur. Phys. J. B 32, 215-225 (2003).
[17] M. Nicodemi, A. Fierro, A. Coniglio, Europhys. Lett. 60, 684 (2002). A. Fierro, M. Nicodemi, A. Coniglio, Jour. Phys.: Cond. Matt. 15, S1095 (2003).
[18] M. Tarzia, A. Fierro, M. Nicodemi, and A. Coniglio, in preparation.
[19] M. Tarzia, A. de Candia, A. Fierro, M. Nicodemi, A. Coniglio, Europhys. Lett. 66, 531 (2004).
[20] M. Mézard and G. Parisi, Eur. Phys. J. B 20, 217 (2001).
  G. Biroli and M. Mézard, Phys. Rev. Lett. 88, 025501 (2002).
[21] Along with the fluid solution, also a crystalline solution, characterized by the breakdown of the horizontal translational invariance, can be found. Here we consider the case where the crystallization is avoided, and limit the calculation to the fluid phase.