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Size segregation and convection

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Abstract. – The size segregation of granular materials in a vibrating container is investigated using Molecular Dynamics. We find that the rising of larger particles is accompanied by the existence of convection cells even in the case of the lowest possible frequencies. The convection can, however, also be triggered by the larger particle itself. The possibility of rising through this mechanism strongly depends on the depth of the larger particle.

One of the most puzzling phenomena encountered in granular matter is size segregation: When a mixture of grains of the same material (equal density) but different size is shaken in a container the larger particles rise to the top. This effect has been extensively studied experimentally [1, 2, 3] and has much importance in numerous industrial processes[4]. Recently this so called "Brazil nut effect" has attracted much interest among physicists[5].

Size segregation inevitably seems to contradict equilibrium statistical mechanics since the density of the overall packing increases with polydispersivity and so gravity makes situations with larger particles on the bottom energetically more favourable. Rosato et al.[6] proposed a Monte Carlo algorithm and put forward a kinetic argument to explain segregation using the fact that smaller particles are more mobile. In the same year Haff and Werner[7] did Molecular Dynamics simulations of rather small systems and claimed that segregation was essentially a consequence of solid friction and the rotation of the particles. Jullien et al.[8] used a piling technique which is non–stochastic as compared to the one of ref. 6 and found a critical ratio $\mathcal{R}$ for the radia of spherical particles below which no segregation occurs. Based on these ideas Duran et al.[9] formulated a geometrical theory for segregation in which the small particles glide down along the surface of the larger particle, the critical ratio $\mathcal{R}$ of radia being between continuous and discontinuous gliding. They also claimed experimental evidence for two types of dynamics and visualized the discontinuous ascent of the larger particle through stroboscopic photos. Jullien et al.[10] reproduced the discontinuous dynamics by including horizontal random fluctuations into their model.

Parallel to these local theories there has been the "convection connection": It is known experimentally[11] and numerically[12, 13] that shaken assemblies of spheres form convection rolls which are attached to the walls of the container. For weak shaking the convection rolls only appear on the surface. Knight et al.[14] showed experimentally that segregation was due to this convection and the fact that larger particles have a harder time entering again into the bulk once they are on the surface. They also verified an exponential decay of the convection strength as function of depth for weak shaking[15]. Duran et al.[16] verified segregation due to convection in two dimensions for strong shaking and claimed that above mentioned local mechanisms are at work at weak shaking. Their stroboscopic pictures showed convection cells above the particles.

Here we present large scale Molecular Dynamics simulations showing that also for weak shaking convection is responsible for segregation but in a more intricate way: Under certain conditions the larger particle is able to pull down the convection rolls due to the more efficient momentum transfer and then rises within its flow. Because of the exponential decay of convection with depth the ability to rise critically depends on the vertical position of the larger particles.

We used the classical Gear predictor–corrector Molecular Dynamics[17] algorithm and the contact forces proposed by Cundall and Strack[18]: Two grains i and j at positions $\vec{r}_i$ and $\vec{r}_j$ interact if the distance between the center points is smaller than the sum of their radii $\mid \vec{r}_i - \vec{r}_j \mid \; < \; R_i + R_j$. For this case the force between particles i and j moving with the velocities $\dot{\vec{r}}_i$ and $\dot{\vec{r}}_j$ and rotating with angular velocities $\dot{\Omega}_i$ and $\dot{\Omega}_j$ is given by

$$\vec{F}_{ij} = F_{ij}^N \cdot \frac{\vec{r}_i - \vec{r}_j}{|\vec{r}_i - \vec{r}_j|} + F_{ij}^S \cdot \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix} \cdot \frac{\vec{r}_i - \vec{r}_j}{|\vec{r}_i - \vec{r}_j|} \; . \tag{1}$$

with the normal and shear forces being

$$F_{ij}^N = Y \cdot (R_i + R_j - |\vec{r}_i - \vec{r}_j|) + \; \gamma_N \cdot m_{ij}^{eff} \cdot |\dot{\vec{r}}_i - \dot{\vec{r}}_j| \tag{2}$$

$$F_{ij}^S = sign\left(|\vec{v}_{ij}^{rel}|\right) \min \left\{ \gamma_S \cdot m_{ij}^{eff} \cdot |\vec{v}_{ij}^{rel}| \; , \; \mu \cdot |F_{ij}^N| \right\} \; . \tag{3}$$

The relative velocity $\vec{v}_{ij}^{rel}$ between i and j and the effective mass m_{ij}^{eff} of the particles i and j are defined as

$$\vec{v}_{ij}^{rel} = (\dot{\vec{r}}_i - \dot{\vec{r}}_j) + R_i \cdot \dot{\Omega}_i + R_j \cdot \dot{\Omega}_j \tag{4}$$

$$m_{ij}^{eff} = \frac{m_i \cdot m_j}{m_i + m_j} \; . \tag{5}$$

The resulting momenta M_i and M_j acting upon the particles are

$$M_i = F_{ij}^S \cdot R_i \quad , \quad M_j = -F_{ij}^S \cdot R_j \; . \tag{6}$$

Eq. (3) takes into account that the particles slide on each other for the case that the inequality $\mu \cdot \mid F_{ij}^N \mid \; < \; \mid F_{ij}^S \mid$ holds, otherwise they feel some viscous friction.

Throughout our simulation we used the parameters $Y = 3 \cdot 10^6 \; g/sec^2$ (Young modulus), $\gamma_N = 100 \; sec^{-1}$, $\gamma_S = 1 \; sec^{-1}$ (phenomenological normal and shear friction coefficients) and $\mu = 0.5$ (Coulomb friction parameter). We considered $N = 950$ particles with radii uniformly distributed in the interval $R_i \in [0.85, 1.15] \; cm$ and with masses $m_i = 2\pi R_i^2 \rho$, $\rho = 1 \; g/cm^2$. The particles are put into a two–dimensional box having walls made of particles with the same material characteristics as the grains and which vibrates vertically according to $y_{box} = A \cdot \sin(2\pi f t)$ with $A = 2 \; cm$. Gravity acts in negative y–direction $F_i^{gr} = -m_i \; g$, $g = 981 cm/sec^2$. The time step for the numerical integration of the Gear predictor–corrector scheme of 5th order was $\Delta t = 5 \cdot 10^{-5} sec$.

We investigate segregation and convection behaviour as function of the vibration frequency f in two different systems, either all particles are small ($R_i \in [0.85, 1.15]$ cm) or we add one single big particle of $R_1 = 4$ cm located in the centre of the box close to the bottom. In order to investigate closer what happens at the onset of segregation we keep all the other parameters fixed. Fig. 1 shows the convection cells without the larger particle (left) and with the larger particle (right) for three different frequencies $f = 2.6$ sec^{-1} (upper figure), $f = 2.8$ sec^{-1} (central figure) and $f = 3$ sec^{-1} (lower figure). For $f = 2.8$ sec^{-1} the convection cells of both systems, with and without the big particle, differ significantly while they are quite similar in the other cases. This indicates that there is a certain frequency interval where the presence of the big sphere triggers the onset of convection which finally leads to segregation. Indeed we find that convection is always present when segregation happens. It is important to note that if one is close enough to the onset of segregation just putting the larger particles one row lower can entirely supress the effect of segregation. This dependence of segregation on the height is quite strong and has not been discussed in the literature. By changing the frequency f very slowly and measuring the convection flow through a plane at a certain height we observed that the transition from the fluctuation regime to the convection regime is very sharp and differs between different runs for less than $\Delta f = 0.05$ sec^{-1}. Moreover when increasing the frequency the transition occurs almost exactly at the same frequency as when decreasing the frequency, i.e. there is no hysteresis. This is in contrast to the case of viscous fluids which tuned out of equilibrium by applying a temperature gradient have nonequilibrium dissipative structures, e.g. Bénard convection, with a characteristic hysteresis.

The triggering of convection cells by the big particle is investigated more quantitatively by calculating the convective flux Φ defined as the sum of material (mass) flow in the centre of the box j_{top} and the flow close to the walls j_{bot} by considering that these flows have opposite signs as illustrated in Fig. 2. The flows j_{top} and j_{bot} are defined by adding the number of particles which move in one direction minus the ones moving in the opposite direction and is measured in particles per cycle time of the box. In fact we measure for each particle if the positions at subsequent nodes of the vibration are on different sides of a height line, where the height of the box was divided into 80 height lines between the bottom of the box and the surface of the packing. Fig. 3 shows the convective flux Φ through planes at different heights D for both systems and for the three different frequencies. For $f = 2.6$ sec^{-1} we find almost no directed flow but only fluctuations. For $f = 3$ sec^{-1} both systems behave similarly, as could also be observed in Fig. 1. For $f = 2.8$ sec^{-1}, however, the convection cells clearly extend deeper due to the existence of the larger particle. Apparently the larger particle is able to pull the convection cells down.

This effect can be explained by the fact that in the region around the large particle the accelerations are higher since the momentum is transferred with less dissipative loss through the larger particle than through a corresponding pile of smaller particles of the same volume. We measured the sum of the absolute values of the forces of all the particles in a region around the position of the large particle and averaged it over time. The region was ring shaped with the inner border of radius 4.5 cm and the outer border of radius 8 cm. At the onset of segregation ($f = 2.8$ sec^{-1}) the average force of the small particles around the large particle is about 15% larger than that of the small particles in the same region in a system containing no large particle. This effect is strongest in the lower part of the ring shaped region. For $f = 2.6$ sec^{-1} the difference is only 5%. Therefore the accelerations in the region around a large particle are larger than if no particle would be present. We believe that this increase in high frequency oscillations is responsible for pulling the convection rolls down. It is, however, interesting to note that the granular temperatures in this region is roughly the same in the two systems.

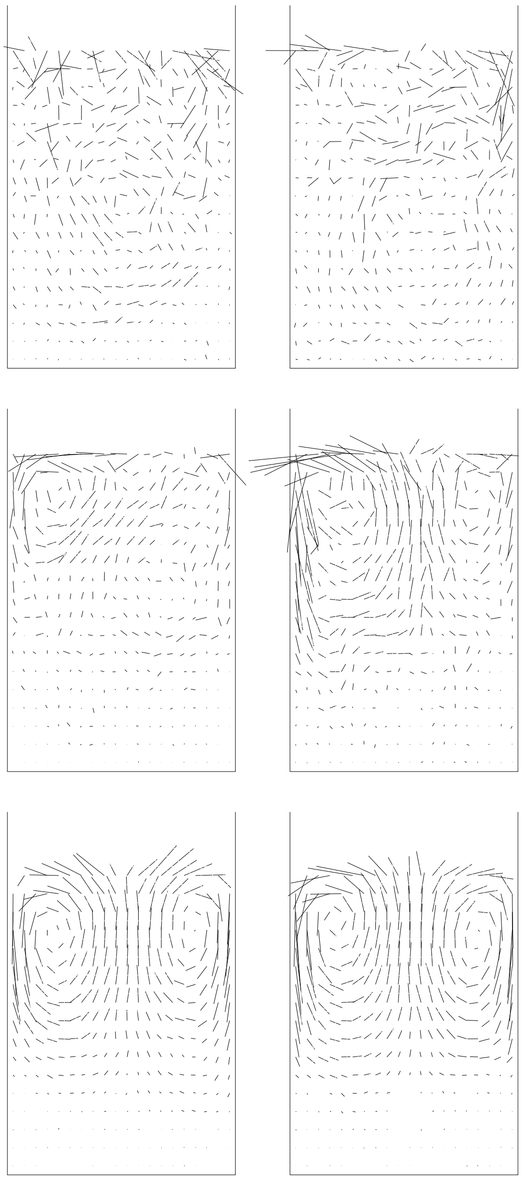

Fig. 1. – Convection rolls in systems without (left) and with the larger particle (right) for three different shaking frequencies, $f = 2.6\ sec^{-1}$ (upper figure), $f = 2.8\ sec^{-1}$ (central figure) and $f = 3\ sec^{-1}$ (lower figure). In the central case the big particle triggers convection rolls. The pictures were obtained by averaging over 50 shaking periods of the box.

One can see from Fig. 1 that the convection cells decay very sharply in strength but that even in the deep regions some essentially horizontal motion occurs. This is reminiscent of the stroboscopic pictures of [16] implying that even in the low acceleration regime some particles move inward horizontally. Within our framework this motion could be interpreted as the exponentially weak tail of the convection rolls.

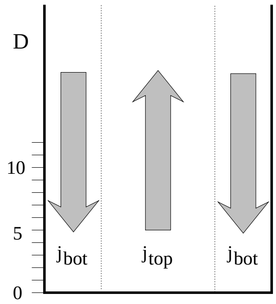

Fig. 2. – Schematic diagramm illustrating how the material flow j_{bot} close by the walls and j_{top} in the center were added to obtain the convective flow Φ.

Next we investigated the dependence of the onset of convection on the ratio of radia $\mathcal{R}$. Note that in our case $\mathcal{R} = R_1/1cm$ because the mean radius of the small particles is $1cm$. For $f = 3.2\ sec^{-1}$ we observed a big particle of radius $\mathcal{R} = 4.0$ moving up immediately. We also studied the cases $\mathcal{R} = 3.5$, $\mathcal{R} = 3.0$, $\mathcal{R} = 2.5$ and $\mathcal{R} = 2.0$. In these cases the large particle remains a certain waiting time on the bottom before it suddenly moves up quite rapidly. Fig. 4 shows a typical evolution of the vertical position of the big particle with radius $\mathcal{R} = 2.0$ and $\mathcal{R} = 3.0$. The waiting times do not noticeably depend on $\mathcal{R}$ being of the order of 30 *sec*. Once the large particle comes to the top it performs an oscillating motion going up and down (whale effect) that has also been observed experimentally[19]. This motion seems due to the convection rolls: In the case where $\mathcal{R}$ is smaller the oscillating motion is more regular because the larger particle has less difficulty reentering into the bulk from the surface and to follow the convective motion. For larger $\mathcal{R}$ the larger particle has more difficulty reentering thus leading to a more erratic horizontal motion as seen in Fig. 4. Particles with smaller $\mathcal{R}$ also seem to dip deeper into the bulk showing that the convection cells can move them more efficiently.

We have shown with large numerical calculations of granular media in a vibrating box that in two dimensions segregation is intimately connected to convection. The larger particles, surrounded by a region of higher acceleration, loosen the material thus deepening the penetration of the convection rolls from the top into the medium. After some waiting time the larger particles are caught by the lower part of the rolls and pulled up. This triggering effect is only relevant close to the characteristic onset frequency of segregation which is sharply defined and strongly dependent on the initial depth of the large particle. Once the large particle is on the top it periodically goes up and down (whale effect) driven by the convection cells.

In addition to convection arching and geometry are also important as pointed out by many authors in the past. How the large particles move in the exponentially weak convection field before being lifted upwards and the $\mathcal{R}$-dependence of the mobility are probably best described by Duran et al.'s local arching mechanisms[9, 16]. The fact that the whale effect of larger particles is less pronounced is certainly due to their lower mobility because of steric hindrance effects as formulated by Rosato et al.[6].

Many of the details of segregation are still not completely clear and in particular in three dimensions additional geometrical effects might play a role. This as well as other questions are difficult to study conclusively with our numerical technique due to the excessive requirements in computer time. It would for instance be interesting to see what happens when the box is so wide that the walls of the box are much farther away from the large particle than the height of the packing. In this experimentally relevant case the walls would not be able to stabilize

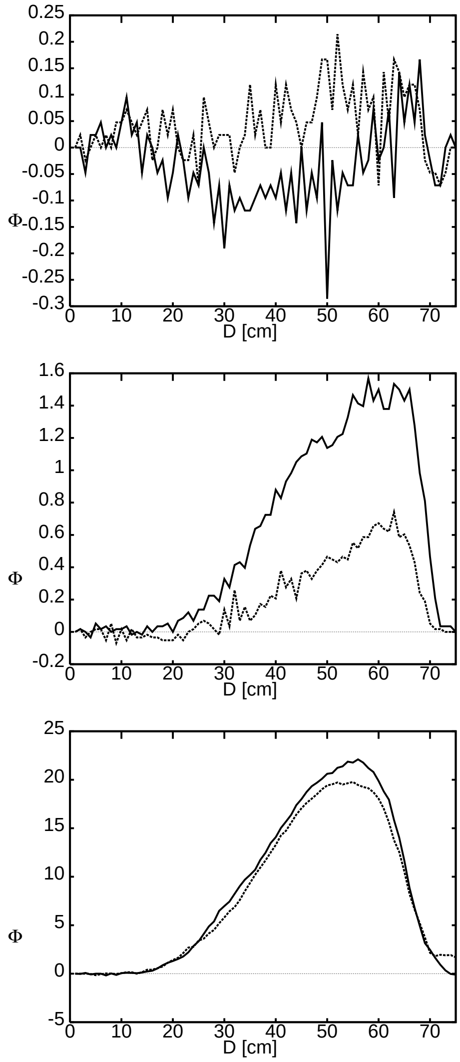

Fig. 3. – Strength of the convection rolls measured through the flux Φ as a function of the height D, $f = 2.6\ sec^{-1}$ (upper figure), $f = 2.8\ sec^{-1}$ (central figure) and $f = 3\ sec^{-1}$ (lower figure). The height D is measured such that the bottom is at the origin of the axis. The flux Φ is measured in units of particles per period. In the central case the convection rolls are stronger and reach deeper inside the material in the presence of the big particle. This triggered convection roll catches the big particle and forces it to rise to the top.

the convection rolls. Simulations with periodic boundary conditions have, however, provided rather similar results as with fixed boundaries [20]. It would also be interesting to study larger ratios $\mathcal{R}$ in order to verify predictions made about a characteristic value of $\mathcal{R} = 12$ [8, 9] but for that case one would need to consider substantially larger systems. The limitations in observation time due to the computational requirements also puts limits on the determination of the segregation velocity and we cannot exclude that particles rise on time scales much larger than the ones accessible numerically.

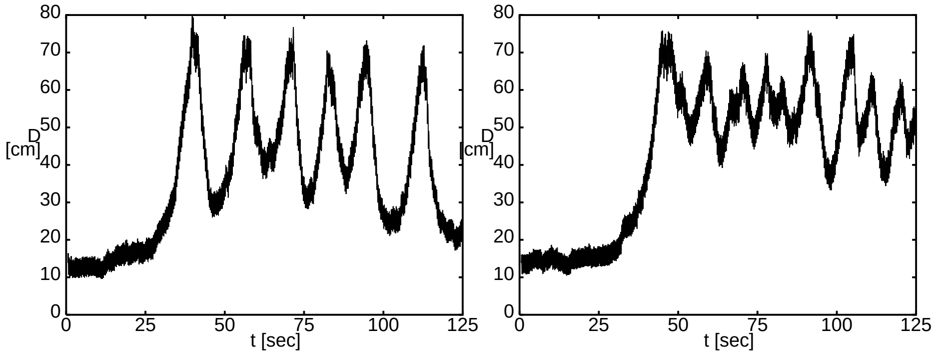

Fig. 4. – Evolution of the vertical position of the big particle as function of time, (a) $\mathcal{R} = 2.0$ and (b) $\mathcal{R} = 3.0$ for $f = 3.2\ sec^{-1}$.

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