The timescales of granular segregation in horizontally shaken monolayers

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The results of an experimental investigation of segregation phenomenon in thin layers of a binary multi-particle system on a horizontal vibrating tray are discussed. Complex structures are observed to emerge from initially mixed states and result from interaction of individual particles. Qualitatively different segregation states are found which have disparate timescales and these are shown to have systematic dependence on the control parameters.

Key words: dry binary monolayer, segregation, horizontal vibrations, coarsening, multiple timescales.

1. Introduction

Dry granular materials can be considered as large collections of individual particle whose collective macroscopic properties can be quite different from those of the familiar forms of the matter, solids, liquids, or gases (Jaeger et al., 1996). The main forces acting on individual particles are inelastic collisions and friction and are inherently dissipative. Also, the individual particles are too large for thermal fluctuations to play a role in a granular system. Therefore, any dynamical process involving grains requires a significant input of energy which is typically supplied in the form of external vibration or shear.

A counterintuitive feature of granular matter is segregation, which is observed when an initially mixed state evolves into one where constituent components of the mixture separate to form clusters. The phenomenon can result from differences in size, rigidity, density, or surface properties of particles (Williams, 1976; Rosato et al., 1987; Mullin, 2002). Segregation provides a striking example of the emergence of order in a complex system, since patterns can form spontaneously in shaken mixtures of dry cohesionless particles. However segregation does not always happen and predicting the conditions for its occurrence is far from trivial. Despite significant research effort, the processes that give rise to segregation are yet to be fully described and understood (Aranson and Tsimring, 2006).

Segregation is not only of deep scientific interest, it is also of practical relevance in fields, such as agriculture (Gillis, 1958; Clementson et al., 2009), geophysics (Werner and Hallet, 1993; Kessler and Werner, 2003; Rosato and Blackmore, 1999; Johnson et al., 2012) and engineering (Fan et al., 1990; Engblom et al.,...
The primary objective in many industrial processes is to achieve complete particle mixing, rather than an undesired separation and clustering of particles (Williams, 1976; Barbosa-Cánovas et al., 2005). Applications range from food preparation to the creation of ceramics and beauty products and drugs. The rich variety of industrial problems and natural phenomena has stimulated a number of controlled small scale laboratory experiments, at first in engineering disciplines, and more recently in the physics community. Segregation has been reported in vertically and horizontally shaken beds (Ahmad and Smalley, 1973; Shinbrot and Muzzio, 2001), rotating drums (Seiden and Thomas, 2011), emptying and filling of vessels (Shinohara, 2006), avalanching (Wiederseiner et al., 2011; Gray, 2010) and chute (Fan and Hill, 2011a,b) flows.

The particular system which will be the focus of attention here was first studied by Mullin (2000), who showed the pattern formation in binary monolayer of particles on a horizontal rectangular tray subject to a periodic shaking (see Fig. 3a for the schematics of the experimental setup and §2 for its description). Both types of particles lay on the surface of the tray and were driven by stick-slip interaction with it. This resulted in anisotropic random motion of the individual particles in two dimensions with a preference for motion in the direction of the forcing. The particles responded to the driving differently because of the differences in their mass, shape and surface properties. Mullin (2000) reported a continuous, but slow time-evolution of the patterns on the timescale of hours (see Fig. 1) that followed $t^{1/4}$ power law, in agreement with a model problem developed for the geological phenomenon of ‘stone striping’ by Mulheran (1994).

Following this study, Reis and Mullin (2002) showed that an initially homogeneous mixed layer of particles demixed on the timescale of minutes by forming spontaneous clusters of its constituent components if the combined filling fraction of the layer ($C$) was increased beyond a critical value ($C_{cr}$). At low values of $C$ the material remained mixed and the positions of the particles were disordered. At intermediate values of $C$ mobile liquid-like clusters of particles were observed, whereas at high values of $C$ stripes formed in a direction orthogonal to the drive. The three phases of segregation were termed binary gas (unsegregated, see Fig. 4a), segregation liquid (mobile clusters, see Fig. 4b) and segregation crystal (stripes, see Fig. 4c), respectively (Reis et al., 2004). Thus far it is unclear how these short timescale findings relate to the observations by Mullin (2000). Indeed, one might anticipate that for sufficiently long times patterns reported by Reis and Mullin (2002) disappear. One of the aims of this paper it to seek possible links between the two sets of results.

Many attempts have been made to model the experiments in horizontally shaken binary monolayers of dry particles (Aranson and Tsimring, 2006). For example, Pica Ciamarra et al. (2007) have suggested that the segregation is a result of a dynamical instability which resembles the classical Kelvin-Helmholtz waves observed at a fluid interface. Stripe formation similar to that in the short timescale experiments of Reis and Mullin (2002) was also observed in models for binary fluids in which the components are differentially forced (Pooley and Yeomans, 2004; Sánchez et al., 2004). In this class of system the differential drive between each of the particle species, which can be interpreted as different frictional interactions of two types of grains with the surface of the oscillatory tray, is the principle driving mechanism for segregation. However, the segregation by friction
on its own is not sufficient to always explain the behaviour of particles on the oscillating plate as shown experimentally by Kondic et al. (2003).

By adding artificially high levels of noise to all particles in a MD simulation of this experiment, Ehrhardt et al. (2005) have shown that it is possible to create a threshold in $C$, above which the segregation appears. Furthermore, a number of numerical investigations predicted switching between the horizontal and vertical stripes in the segregated state either by changing the amplitude and frequency of the driving and the tray size (Shi and Ma, 2005), or by adjusting the relative filling fraction of different types of particles (Fujii et al., 2012). Interestingly, switching of direction of the patterns was never observed experimentally and both Mullin (2000) and Reis and Mullin (2002) reported segregation in form of stripes perpendicular to the direction of shaking.

Here we present results of an extensive experimental investigation into the short and long timescale segregation processes in binary mixture on an oscillating horizontal tray. We establish connections between the apparently contradictory behaviours reported by Mullin (2000) and Reis and Mullin (2002) by repeating the experiments in the same parameter regime as the previous studies. Specifically, we establish that the coarsening, reported by Mullin (2000), takes place in the segregation liquid phase. Moreover, we show that reduction in the amplitude of the oscillations of the tray leads to long timescale coarsening that takes place regardless of short timescale behaviour. The coarsening is independent of the total filling fraction and results in a qualitatively different segregation pattern. More specifically, we observed that if the amplitude of the oscillations is reduced the stripes form parallel to the direction of shaking as predicted by Shi and Ma (2005).

2. Experiment

In order to study segregation in a binary monolayer of particles, we define the total filling fraction $C$ of the granular layer as

$$C = \phi_1 + \phi_2, \quad \phi_1 = \frac{N_1 A_1}{xy}, \quad \phi_2 = \frac{N_2 A_2}{xy}, \quad \text{(2.1)}$$

where $N_1$ and $N_2$ are the numbers of particles of the respective species of the monolayer, $A_1$ and $A_2$ are the corresponding projected two-dimensional areas of the particles and $x$ and $y$ are the longitudinal and transverse dimensions of the rectangular tray. Hence $\phi_1$ and $\phi_2$ are the individual filling fractions of each of the constituents.

We performed experiments using binary mixtures of phosphor-bronze spheres (with diameter $d = 1.5$ mm, polydispersity $p = 2\%$ and density $\rho = 8.96$ g cm$^{-3}$) and rough poppy seeds which have shape of an artist’s palette (average projected surface area of $A_2 = 0.86$ mm$^2$, $p = 15\%$ and $\rho = 0.2$ g cm$^{-3}$).

We also did experiments with other mixtures of particles which had significantly different properties to poppy seeds and phosphor-bronze spheres, for example spherical and rhomboidal glass particles. Segregation was found in a number of mixtures as shown in Fig. 2 (see also data in Fig. 9). It was observed for mixtures of particles with (i) the same density and shape, but different sizes (Fig. 2a, c), (ii) same size and shape, but different densities (phosphor-bronze...
and nylon spheres with projected surface area of 1.98 mm$^2$ were considered by Giannasi (2010), (iii) particles with similar polydispersities (Fig. 2b) and (iv) many mixtures of different combinations of particles (Fig. 2d-h), where particles were typically smoother and heavier than poppy seeds, but lighter and rougher than phosphor-bronze spheres. In fact, any mixture created by a combination of particles in Fig. 2 was also observed to segregate. Therefore, segregation was found to be a very robust phenomenon.

We chose to study the mixture of phosphor-bronze spheres and poppy seeds for a number of practical reasons. Firstly, the poppy seeds were effectively an order of magnitude smaller than the spheres and hence changing their number gave sensitive control of the filling fraction. In all experiments we varied $C$ by keeping the filling fraction of the spheres fixed at $\phi_1 = 0.1931$, although the segregation phenomena were found to be robust to changes in either type of material (Reis et al., 2006). Secondly, the phosphor-bronze spheres have a high reflection coefficient and are hence relatively easy to visualise compared to other materials used (see description of the visualisation procedure). The relative difference in sizes and shapes of the chosen mixture also implied that the segregation process (and, most importantly, the coarsening) was relatively quick compared to other mixtures of particles (see, for example, Giannasi (2010) for the segregation timescales in the mixtures of spherical particles of the same size) and that problems, associated with the maximum packing of monodispersed particles were minimised. Finally, the mixture of phosphor-bronze spheres and poppy seeds has been well studied in the past (Mullin, 2000; Reis and Mullin, 2002; Reis et al., 2004, 2006), which provided us with the necessary background for the current study.

A thin granular layer was used such that there was an approximate monolayer of material on the surface of the tray. By definition the total filling fraction $C$ varies between zero and a value corresponding to the maximum packing in two dimensions. For monodisperse disks the latter is $C_{\text{max}} = \frac{\pi}{\sqrt{12}} \approx 0.9609$. This number is even higher for the binary mixture of disks: by taking the values of $A_1 = 1.77$ and $A_2 = 0.86$, which correspond to projected surface areas of a phosphor-bronze sphere and a poppy seed, respectively, we obtain $C_{\text{max}} \approx 0.9165$ (Uche et al., 2004). Of course, the poppy seeds are highly irregular disks, which in theory means that $C_{\text{max}} \rightarrow 0.991332$: a maximal allowable value of packing fraction for two species of particles. Furthermore, the poppy seeds can remain in contact with the tray, but have the projected surface area less than $A_2$, which was measured using the largest projected surface area of a poppy seed. Therefore, the experiments were also performed with higher values of $C$ than $C_{\text{max}}$. In practice the larger and heavier phosphor-bronze spheres were always in a monolayer, but the lighter and flatter poppy seeds could partially overlap because of both the polydispersity of the poppy seeds and the difference in size and shape between the poppy seeds and the spheres. However, the phosphor-bronze spheres were observed to remain in contact with the surface of the tray and the poppy seeds were on average in contact with the surface of the tray. The approximate monolayer regime ensured a near homogeneous forcing of all particles and enabled the visualisation of the system by taking images from above.

The experimental apparatus in our study was the same as one used by Reis and Mullin (2002) (see the schematic diagram in Fig. 3a) with the exception that the
majority of our experiments were performed in a horizontal rectangular tray with dimensions \((x, y) = 90 \times 90\) mm. We also repeated the experiment in a larger tray of \((x, y) = 180 \times 90\) mm to check that the reproducibility of results did not depend on system size. The tray was connected to a Ling LDS V409 electromechanical shaker, so that it vibrated longitudinally and horizontally with respect to gravity. Its motion was constrained to be unidirectional by four lateral high precision linear bearings and was monitored using a LVDT and an accelerometer. This movement was confirmed to be sinusoidal to better than 1%. The setup was mounted on a heavy base which could be adjusted using fine threaded screws to ensure its horizontal levelling.

First the tray was levelled statically using a high precision engineering spirit which was accurate to better than \(\pm 0.01\)°. Further levelling was performed dynamically by shaking individual species of particles at a fixed amplitude and frequency. Optimal levelling was achieved when the distribution of all particles was spatially isotropic and homogeneous (Kudrolli et al., 1997). The spherical particles were particularly sensitive to any inclinations of the tray, since they rolled very easily. However, on longer timescales poppy seeds also slid down the slope if the tray was not adequately levelled. We found that a satisfactory long-time dynamical levelling was achieved for relatively small magnitude of forcing, so all our experiments were performed at just two amplitudes \(a = \pm 0.6\) mm and \(a = \pm 1.2\) mm at a fixed frequency of \(f = 12\) Hz. This was also favourable from the practical point of view, because the experiments were very time-consuming, each data acquisition taking typically 48 h to complete.

The dimensionless acceleration of the tray can be defined as (Jaeger et al., 1996):

\[
\gamma = \frac{4\pi^2 af^2}{g},
\]

(2.2)

where \(g\) is acceleration due to gravity. We can relate the amplitudes used in this study to the experimental parameters reported by Reis et al. (2006): the two amplitudes correspond to \(\gamma = 0.348\) and \(\gamma = 0.696\), respectively. The parameter \(\gamma\) is commonly used in vertically vibrated granular systems, in which gravity plays a dominant role (Jaeger et al., 1996). However, in our problem of horizontal shaking \(g\) is also present through the frictional forces acting on particles. Moreover, Reis et al. (2006) reported the transition value of the dimensionless acceleration \((\gamma_c)\) between the segregated and mixed phases, suggesting that \(\gamma\) is the appropriate parameter to describe the forcing of the tray.

The segregation process was visualized by uniformly illuminating the tray, and images of patterns were captured from above using either CCD camera (75 frames per second) to obtain short time-scale data, or a Nikon D300S still camera for data for coarsening processes which occurred on timescales of hours. The phosphor-bronze spheres have a higher reflection coefficient and are hence more visible than the poppy seeds, so monitoring their motion was more convenient. Each sphere corresponded to approximately 130 pixels on a high resolution still image. We did not perform tracking on poppy seeds. The position of individual phosphor-bronze spheres was recovered using image processing, developed in MATLAB R2009a and applied to the high resolution still images. All the measurements were taken in a central \((70 \times 70)\) mm visualisation window of the \((90 \times 90)\) mm tray in order
to achieve the necessary resolution of the images to enable good estimates of the centres of the spheres.

The experiments were started from a close approximation to a homogeneous binary mixture with a few exceptions mentioned explicitly later. It was important to initiate the experiments with controlled initial conditions since there were multiple segregated states. Hence seeding the initial state by starting the experiment with clusters could lead to different final states. Repeatable initial conditions were achieved in the following procedure. First the required filling fraction of poppy seeds, $\phi_2$, was vibrated at a large amplitude of $a = \pm 5$ mm until a uniform layer was created. Next the phosphor-bronze spheres were suspended above the layer on a horizontal perforated plate with 2 mm diameter holes arranged on a triangular lattice. The shutter was opened and the spheres were released onto the layer of other material, creating an approximately homogeneous binary mixture. This procedure was time-consuming but it ensured repeatability in the results.

3. Segregation process

(a) Experimental observation on fast timescales

The first set of experiments was aimed at checking the observations of short timescale behaviour, reported by Reis and Mullin (2002) and Reis et al. (2004, 2006). Unless otherwise stated for all the results that follow the mixture was shaken at $\gamma = 0.696$, which corresponds well to the parameter values used in previous investigations. In summary, we confirm the existence of three qualitatively distinct phases of the mixture of phosphor-bronze spheres and poppy seeds, binary gas, segregation liquid and segregation crystal. These formed on the timescale of minutes. The details are given in Figs 4 & 5 and are discussed below.

Quantitative measurements of the segregation process were made using a microscopic measure of the local Voronoi density by taking the following steps. Firstly, we constructed the Voronoi cells for the spheres by tessellation using the voronoi(x,y) routine in the package MATLAB R2009a. An example of such a tessellation is given in Fig. 3b, c. For each $i$ of the $N_1$ phosphor-bronze spheres, where $i \in [1, N_1]$, the procedure yields a polygonal cell that encloses the region of the tray. Any point inside this region is closer to the centre of the $i$th particle than any other sphere on the tray. Once the Voronoi polygons were created, we defined the local Voronoi area density of the $i$th sphere

$$\rho^i = \frac{A_1}{A^i_{cell}},$$

where $A_1 = \pi(d/2)^2$ is the two-dimensional projected area of the sphere with diameter $d$ and $A^i_{cell}$ is the area of its Voronoi polygon. The alternative measures of the segregation, such as the width of the cluster and the particle-particle correlation function were explored elsewhere (see Reis and Mullin (2002) and Reis et al. (2004), respectively) and were found to correspond well to the measure chosen here.

From the definition of the local Voronoi area density it follows that the closer the phosphor-spheres are to each other the closer $\rho^i$ of an individual sphere is to 1.
However, this parameter varies significantly even within the same pattern. For example, the particles at the edges of the segregation clusters had an associated polygon areas which were visibly larger, and hence area density significantly lower than those in the bulk of the domains as can be seen in Fig. 3c. Therefore, we constructed the probability distribution function for the local Voronoi area density of a pattern, $PDF(\rho)$, by constructing normalized histograms of $\rho^i$ as shown in Figs 4d-f & 5a. We built $PDF(\rho)$ once the pattern reached its saturated state, which was found to be 5 minutes after the start of the experiment from a homogeneous and isotropic mixture as in Reis et al. (2006). In practice, patterns remained approximately unchanged for at least another 10 minutes in all cases. Furthermore, a time window of 9.44 s which corresponded to approximately 700 images, was used to obtain statistical averages for local Voronoi area densities. Each $PDF(\rho)$ typically contained statistical ensembles with more than 300,000 particles.

Typical $PDFs$ of local Voronoi area densities for different filling fractions of the tray, which correspond to three phases of segregation, are shown in Fig. 4d-f, respectively. Each of the distributions has two clear peaks, where one is close to $\rho = 0$ and is nearly independent of $C$, and a second peak moves towards $\rho = 1$ as $C$ is increased. The first peak corresponds to the mean local Voronoi area density of particles at the edge of the patterns and the second is the mean value of the local Voronoi area density of particles in the bulk of the clusters. Hence, we define the characteristic local Voronoi area density, $\rho^m$, based on the position of the principle peak of the distribution as illustrated in Fig. 5a. Changing the value of $C$ also affects the width of the distribution. We use a typical deviation $W/2$ from $\rho^m$ as a measure of the fluctuations of the motion of the particles. Without loss of generality, we choose to define the fluctuations of the characteristic local Voronoi area density by evaluating $W$ at $3/4$ of the peak (Fig. 5a).

The variation of $\rho^m$ and $W$ as functions of $C$ is shown in Fig. 5b. As reported by Reis and Mullin (2002) and Reis et al. (2004, 2006), the short-time behaviour of particles indicates a second-order phase transition from a mixed state to one in which the constituents cluster. The characteristic local Voronoi area density changes continuously with increasing $C$ with a peak in the mean amplitude of the fluctuations at the critical point. The boundary between the binary gas and the segregation liquid phases is marked by the continuous phase transition point, $C^{cr} = 0.667 \pm 0.09$, which is in close accord with value reported by Reis and Mullin (2002).

(b) Stability of segregation patterns: short-time vs long-time experiments

The long-time stability of the patterns in Fig. 4a-c was examined by shaking the tray for 48 hours and recording images of the granular material. For all filling fractions that correspond to the binary gas and segregation crystal phases (see Fig. 5b) the final state was formed 5 minutes into the shaking from initially homogeneous state and persisted until the end of the long-time experiment. However, it must be emphasised that mixed initial conditions were crucial for obtaining the reproducible patterns in the segregation crystal phase. Otherwise the stripe width could be biased if the initial state contained large clusters of metal particles.
On the other hand, the outcome of the long-time experiment for the intermediate values of the filling fractions between the two values of $C$ (0.667 ± 0.09 and 0.953 ± 0.22) indicated by the two vertical lines in Fig. 5b showed coarsening in accord with Mullin (2000). The patterns in the liquid phase were highly irregular at first, slowly evolved by merging and passing through a sequence of configurations, until a single stripe was observed in the direction perpendicular to the shaking (Fig. 6, top row). A typical timeline of the pattern evolution in the smaller tray of $(90 \times 90) \text{mm}^2$ was the following (see also Fig. 6, top row): within a minute from the start of the experiment larger clusters of phosphor-spheres formed at random locations. They merged and broke until around 5 minutes into the experiment when metastable structures were formed. At filling fractions close to the boundary between the segregation liquid and the segregation crystal phases the pattern had the form of stripes oriented with their long axes orthogonal to the direction of the vibration. At lower filling fractions the patterns had the form of irregular clusters. This state persisted for approximately one hour, after which further merging was observed and all patterns contained stripes. The coarsening slowed progressively as the stripes broadened. In the small tray the whole process took up to 4 hours depending on the total filling fraction. The final pattern was always a single stripe which was typically located near one of the tray ends. However, both boundaries were chosen with equal likelihood and when formed, the final stripe did not change further. We also repeated the experiment in the larger tray of $(180 \times 90) \text{mm}^2$ with similar results, although the typical timescales for coarsening were on the order of a day. However, multiple stripes distributed along the tray length were also occasionally observed after 48 hours of shaking in the larger tray.

(c) Striping parallel to the forcing

The experimental evidence for the long-time coarsening of the patterns, observed for a range of $C$ values at $\gamma = 0.696$, was also studied by repeating the experiments for $\gamma = 0.348$. In Fig. 6 we show one such comparison, where all the parameters are the same except for the amplitude of forcing of the tray. Both experimental runs result in coarsening, however the final states were qualitatively different. The experiments at lower amplitude revealed a new orientation of the stripe, which was aligned in the direction of shaking. The pattern evolution also slowed because the individual particles moved less rapidly around the tray at the smaller forcing amplitude.

The stripes oriented in the direction parallel to the tray oscillations have not been observed previously. It is feasible that at lower forcing amplitudes a different mechanism is responsible for the segregation compared with the one responsible for the formation of stripes at $\gamma = 0.696$. Interestingly, if the segregation measures are taken after 5 minutes of shaking, the patterns are very similar to those found at larger oscillation amplitudes. The main difference is that clustering appears at lower filling fractions and the resulting patterns have higher wave-numbers. The latter is also visible in a more pronounced first peak of $PDF(\rho)$ because of the larger number of particles at the cluster edges (Fig. 7a-d and Fig. 8, top row).

The long-time segregation is less complicated when $\gamma = 0.348$ than for $\gamma = 0.696$. At lower amplitude we found that the stripe of metal balls always coarsened in the middle of the tray in a direction parallel to the vibration, whereas the poppy
seeds were deposited along the longitudinal walls of the tray, independent of the number of particles on the tray (Fig. 8). In fact, when there were fewer rougher particles on the tray, the bulk segregation was faster, ranging between 2 h for the smallest filling fractions considered \( C = 0.34 \) and 20 h, when the filling fraction of particles was around \( C = 1.2 \). Once the stripe of metal balls was formed, it was stable and persisted for at least 48 h from the start of the experiment.

The coarsening process observed at \( \gamma = 0.348 \) for the filling fractions that correspond to the binary gas phase in Fig. 7e was different compared with all other coarsening behaviours. In this regime the metal balls never formed well-defined clusters. Instead the segregation followed from a gradual displacement of the poppy seeds towards the wall region where there were fewer collision between metal balls. Interestingly, for the filling fractions, that correspond to the segregation crystal phase in Fig. 7e, despite the coarsening process most of the spheres were locally organised into an ordered hexagonal lattice. After 5 minutes of vibration a typical configuration resembled stripes perpendicular to the direction of shaking, which then slowly reoriented themselves as if under rotational shear and merged.

\[(d)\) Measuring coarsening

Quantitative measurements of the coarsening process were made by estimating the mean of the local Voronoi area densities, \( \langle \rho \rangle \), for the phosphor-bronze spheres as the patterns evolved. Results are shown in Fig. 9 for a range of \( C \) and both the amplitudes of oscillations used. The data used to construct the figure were estimated by analysing a single image at a given instant in time, indicated on the abscissa. For each of the data points we found the mean of the distribution, which was calculated from approximately 700 values of the local Voronoi area density. Nevertheless, the chosen measure was found to vary consistently in time \( t \) over the whole range of parameters.

In order to accommodate the long time-scales involved, it proved to be convenient to plot the variation of \( \langle \rho \rangle \) with \( t \) on a log-log scale. All data sets in Fig. 9 are parallel to one another. Using Levenberg-Marquardt algorithm, we fit a power law of the form \( \langle \rho \rangle \sim c(t - t_0)^n + \langle \rho \rangle_0 \), where \( \langle \rho \rangle_0 \) is the mean local Voronoi area density at the start of the experiment, when \( t = t_0 \), and \( c \) is a constant that presumably depends on \( C, a, f \) and the whole range of other system parameters. By fitting this to the experimental points, we obtain \( n \approx 1/4 \), found previously by Mullin (2000) for \( \gamma = 0.58 \) and predicted by a model problem developed for the geological phenomenon of ‘stone striping’ (Werner and Hallet, 1993; Mulheran, 1994). The data is noisier and the curves are shifted towards lower values of \( \langle \rho \rangle \) as the total amount of the material in the system is reduced, because there is more free space on the tray for the phosphor-bronze balls to move around.

We also repeated the coarsening experiments with selected mixtures of particles from Fig. 2, which to a good approximation gave \( \langle \rho \rangle \sim t^{1/4} \) scaling (see Fig. 9). Therefore, despite a relatively large range of parameters used in this study of coarsening, the findings suggest that a form of diffusion, similar to the diffusion in the ‘stone striping’ problem, takes place in the system on the long timescale.
4. Conclusion

Results of an experimental investigation of segregation behaviour in a monolayer of two types of particles placed on a horizontal oscillating tray have been presented. The findings clearly demonstrate that the segregation process has multiple timescales associated with it. Critical behaviour and three qualitatively distinct phases were found on short timescales, as reported previously by Reis and Mullin (2002). However, in the appropriate region of parameter space on the timescale of hours the patterns coarsened to form a stripe in accordance with findings of Mullin (2000).

The experiments were performed at two oscillation amplitudes of the tray. It remains to be established if similar behaviours will be observed at different amplitudes, but previous findings of Reis et al. (2006) suggest that one might expect a systematic dependence of the segregation process on the forcing of the tray. We observed coarsening for other mixtures of particles (see Figs 2 & 9), although the results reported here are mainly for the mixture of poppy seeds and phosphor-bronze spheres. All coarsening experiments that we performed yielded the same $\langle \rho \rangle \sim t^{1/4}$ behaviour. In current theoretical models this suggest a range of potential segregation mechanisms, but a complete theory, capable of capturing all behaviours observed in the experiments, is still missing. Our experimental study will undoubtedly stimulate new theoretical investigations.

A promising start has been made using a cellular-automaton model, similar to Fitt and Wilmott (1992) and developed recently in the University of Manchester (private communication with T. Barker and L. Cawthorne). This discrete model predicts binary-gas-like behaviour for small $C$, coarsening at intermediate values of $C$ and striping caused by clogging of smaller particles as $C$ is increased further, much like that found in our experiments for $\gamma = 0.696$. These finding suggest that both the relative size of the particles of different species and their differential response to the forcing of the tray are responsible for the segregation in our system. Further investigation with the model may reveal other features of the experiment, such as reorientation of the pattern at lower amplitudes of the driving.

An interesting and novel experimental observation is the reorientation of the coarsened pattern with respect to the direction of the shaking when the amplitude of oscillations is small, which agrees qualitatively with theoretical predictions of Shi and Ma (2005). The robustness of this result was checked multiple times by considering the evolution of a stripe, which was artificially created as the initial condition at $C = 0.83$ and $\gamma = 0.348$. The stripe was initially orthogonal to the drive but evolved to align with the forcing through an interesting intermediate state as shown in Fig. 10. All of the behaviours reported here are beautiful examples of how complexity at small scales impacts the emergent order of pattern formation at the macro-level.

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References


Figure 1. Evolution of the pattern for the copper ball/poppy seed mixture vibrated at ±1 mm and 12.8 Hz in the direction orthogonal to the stripes and indicated with the arrows. The images were taken at 5 min, 15 min and 6 h. From Mullin (2000).
Figure 2. Segregation of binary mixture of: (a, c) spherical glass particles, (b) rough carborundum and glass particles, (d, e, g) phosphor-bronze spheres and spherical glass particles, (f) sugar and glass spherical particles, (h) phosphor-bronze spheres and rhomboidal glass particles. Image (e) shows an example of the long-time coarsening of the mixture of spherical glass particles and phosphor-bronze spheres in image (d). Coarsened pattern also shown in (h). The particles in the images are the following: ‘deco’ glass spherical particles of density $\rho = 2.5 \text{ g cm}^{-3}$ and (c) $d = 0.33 \text{ mm, } p = 6\%$, (a, c, f, g) $d = 0.53 \text{ mm, } p = 6\%$, (d, e) $d = 0.87 \text{ mm, } p = 12.5\%$, (a) $d = 1.125 \text{ mm, } p = 11\%$; (b) natural SiC carborundum of density $\rho = 3.21 \text{ g cm}^{-3}$ and size $d = 0.36 \text{ mm, } p = 17\%$; (b, h) ‘potters’ glass particles of density $\rho = 2.5 \text{ g}$ and size $d = 0.45 \text{ mm, } p = 14\%$; (f) sugar spheres of density $\rho = 1.59 \text{ g cm}^{-3}$ and size $d \approx 1.5 \text{ mm}$; (d, e, g, h) phosphor-bronze spheres (properties given in §2).
Figure 3. (a) Schematic diagram of the experimental apparatus. (b) Typical segregation pattern of the granular mixture of poppy seeds and phosphor-bronze spheres for $C = 0.91$, $a = \pm 0.6 \text{ mm}$ and $f = 12 \text{ Hz}$ in the $(90 \times 90) \text{ mm}^2$ tray. The image was taken 5 minutes after the start of the experiment from an initially homogeneous mixture. The pattern is formed in the direction perpendicular to the direction of shaking, shown by the arrow. The phosphor-bronze spheres appear as the bright regions in an image, whereas the poppy seeds together with grain-free areas correspond to the dark regions of an image. (c) An example of Voronoi diagram obtained from the positions of the phosphor-bronze spheres for the mixture shown in (b).
Figure 4. Typical patterns observed in the binary mixture of poppy seeds and phosphor-bronze spheres after 5 minutes of vibration from initially homogeneous mixture at $\gamma = 0.696$ in the $(90 \times 90) \text{mm}^2$ tray and the corresponding probability density function (PDF) of local Voronoi area density, calculated from the positions of phosphor-bronze spheres for (a, d) $C = 0.4528$ (binary gas phase), (b, e) $C = 0.7516$ (segregation liquid phase), (c, f) $C = 1.2188$ (segregation crystal phase). Images correspond to a central $(70 \times 70) \text{mm}^2$ visualisation window. A time window of 9.44 s (which corresponds to $\sim 700$ images) was used to obtain statistical averages for area densities of spheres in three different phases. Each PDF typically contained statistical ensembles which corresponds to $\sim 300,000$ particles.
Figure 5. (a) Typical experimental PDF of local Voronoi area density $\rho_v$ in the binary gas phase, for $C = 0.5824$ and the rest of the parameters as in Fig. 4. The peak in the PDF corresponds to the characteristic local Voronoi density, $\rho_v^m$. The width of the distribution at 3/4 of the peak, $W$, was taken as the measure of the fluctuations of the characteristic local Voronoi density. (b) Dependence of $\rho_v^m$ (left axis, squares) and $W$ (right axis, circles) on the total filling fraction $C$. The curves are drawn to guide the eye. In the region between the solid vertical line and the dashed vertical line, the pattern coarsened if the tray was vibrated for a sufficiently long time (see Fig. 6, top row). For other values of $C$ the patterns were stable after 5 minutes and persisted for at least 48 hours.

Figure 6. Snapshots of evolving patterns in the liquid phase of the poppy seeds/phosphor-bronze spheres mixture, vibrated from an initially homogeneous mixture at $\gamma = 0.696$ (top row) and $\gamma = 0.348$ (bottom row). The time which passed from the start of the experiment is shown in the bottom left corner of each of the images. In both sets of images $C = 0.83$. The snapshots show a central $(70 \times 70)$ mm$^2$ visualisation window of the $(90 \times 90)$ mm$^2$ tray. In the top row the stripes are orthogonal to the direction of shaking, whereas in the bottom row they are parallel to it. The patterns in the final column were stable and persisted for at least 48 hours.
Figure 7. Probability density function (PDF) of local Voronoi area density, $\rho_v$, calculated from the positions of phosphor-bronze spheres 5 minutes after the start of the experiment. The mixture of poppy seeds and phosphor-bronze spheres was vibrated at $\gamma = 0.348$ in the $(90 \times 90) \text{mm}^2$ tray, and the total filling fraction was equal to (a) $C = 0.39$ (binary gas), (b) $C = 0.63$ (segregation liquid) and (c) $C = 1.2$ (segregation crystal). Typical patterns, that correspond to these PDFs, are shown in the top row of Fig. 8. PDFs were constructed in the same way as used in Fig. 4. (d) Typical experimental PDF of local Voronoi area density, $\rho_v$, in the segregation liquid phase, for $C = 0.72$ and the rest of the parameters as in (a)-(f). Two clear peaks emerge for all filling fractions in the segregation liquid and the segregation crystal phases, the first one being fixed around $\rho_v = 0.17$ and the second moving towards $\rho_v = 1$ for increasing $C$. The characteristic local Voronoi density, $\rho^m_v$, and the width of the distribution, $W$, are extracted based on the second peak. (e) Dependence of $\rho^m_v$ (left axis, squares) and $W$ (right axis, circles) on the total filling fraction $C$. As in Fig. 6, the curves are drawn to guide eyes. The solid vertical line corresponds to the boundary between binary gas and segregation liquid phases, whereas the vertical dashed line defines transition between the segregation liquid and the segregation crystal.
Figure 8. Top row: Typical patterns observed in the binary mixture of poppy seeds and phosphor-bronze spheres after 5 minutes of vibration from initially homogeneous mixture at $\gamma = 0.348$ in the (90 $\times$ 90) mm$^2$ tray for $C = 0.34$ (binary gas phase), $C = 0.77$ (segregation liquid phase), $C = 0.91$ (segregation crystal phase). Bottom row: The bulk segregation of the mixture of poppy seeds/phosphor-bronze spheres when the tray was shaken for sufficiently long time. The two images in each of the columns were taken in the same run of the experiment.
Figure 9. Time-evolution of the average local Voronoi density, $\langle \rho \rangle$, plotted on logarithmic scale for a range of experiments. The respective total filling fractions $C$ are indicated in the plot legend. Unless ‘v’, ‘p’ and ‘*’ appear in the the legend, the experiments were performed at $\gamma = 0.348$ in the (90 $\times$ 90) mm$^2$ tray. ‘*’ is used to indicate experiments performed in the bigger tray of (180 $\times$ 90) mm$^2$; ‘v’ indicates experiments carried out with $\gamma = 0.696$; ‘p’ stands for the experiments with the mixture in Fig. 2h. In the above, $\langle \rho \rangle$ was constructed by taking the mean of the Local Voronoi Densities for the phosphor-bronze spheres for each respective time. Power law fits through the data points give powers $n$, indicated in the legend.

Figure 10. A sequence of images of the evolution of the pattern for the poppy seed/phosphor-bronze spheres mixture ($C = 0.83$), vibrated from left to right at $\gamma = 0.348$ in the (90 $\times$ 90) mm$^2$ tray. The time passed from the start of the experiment is indicated in the bottom left corner of each of the images.