



Self-diffusion, local clustering and global segregation in binary granular systems: The role of system geometry



C.R.K. Windows-Yule^{*}, D.J. Parker

School of Physics and Astronomy, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom

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ABSTRACT

Using positron emission particle tracking (PEPT), the influence of system geometry on the dynamical and segregative properties of a three dimensional, vibrofluidised granular system is explored. A relationship between the aspect ratio of a system and the degree of segregation observed is established, demonstrating the ability to adjust segregation intensity within a system simply by modifying its aspect ratio without altering any other parameters, including the number of particles within the system, its volume and the manner in which it is driven. Such a relationship is potentially of value in future research, as well as in industrial settings. The range over which this relationship is valid is determined in terms of the quantity q , the ratio of heating to cooling events within an excited granular system. Within this range, a relationship between self-diffusion, D , and aspect ratio, A , of the form $D = \alpha \exp(\beta A^2)$ is also proposed. Finally, this study presents what is believed to be the first experimental evidence of *sudden chain energy transfer events*, a phenomenon recently discovered in two-dimensional granular beds, in a three-dimensional system.

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1. Introduction

Granular materials – randomly packed aggregates of individually solid particles – play a variety of important roles in nature, industry and everyday life. They are distinct from ‘ordinary’ molecular materials due largely to two defining properties – their macroscopic size, which renders them effectively athermal [1], and the inherently dissipative interactions between components of a granular system [2]. It is this latter property which gives rise to many of the unique and interesting behaviours observed in granular materials, such as convection [3], pattern formation [4] and the spontaneous development of gradients in density and particle energy [5]. Perhaps one of the most striking, but also industrially relevant, phenomena observed is that of *granular segregation*; when a granular system composed of two or more particle species differing in size or material properties is exposed to an energy source, e.g. through contact with a solid vibrating surface, a spontaneous separation of the system into its constituent parts may be observed [6]. This separation may be complete or partial [7] and may occur vertically or horizontally, depending on the system parameters [8].

The presence of, and degree of, segregation occurring in a granular material can be affected by a number of parameters including, for example, the relative densities of the various particle species [9], the strength of the excitation with which the system is driven [10] or the number of particles, N , in the system [11]. However, in various industrial settings where

segregation is important, it may be undesirable or even impossible to vary the aforementioned parameters – a variation of N may affect the output rate of a product; varying the density ratio would require altering the product in question, which is likely to pose significant problems; altering the driving of the system may also be near-impossible, for instance if the excitation is due to vibrations during transport. This study investigates, using fixed values of N , density ratio and excitation strength, the effect of the aspect ratio, A , of a system on the segregative behaviour of a vertically vibrated granular bed. Here, the aspect ratio is determined as $A = H/L$, where H is the resting height of the randomly packed bed and L is the width of the container within which the bed is housed. The investigation aims to allow a better understanding of how to control of segregation simply by changing the geometry of the container within which a granular bed is housed, without necessitating the undesirable variation of other parameters. We determine the range of parameter space over which a simple, monotonic relationship between aspect ratio and *segregation intensity* (a measure of the degree of mixing or separation in a binary system), can be observed, and explore the complex and interesting behaviours leading to the breakdown of this relationship in the most highly constrained systems. It should be noted that although A itself does not *directly influence* the behaviour of a system (rather, the corresponding alterations in system density, collision rate, diffusivity etc. cause the observed changes) the authors choose to use it as the main variable since, unlike these other parameters, it can be directly controlled. As such, its use in this study more easily facilitates the utilisation of the current findings in future research or industrial applications.

^{*} Corresponding author.

E-mail address: windowsyule@gmail.com (C.R.K. Windows-Yule).

2. System details

2.1. Experimental set-up and data acquisition

The experimental system consists of $N = 1000$ 5 mm spherical particles, of which $N_L = 500$ are 'light' particles composed of either glass or aluminium and $N_H = 500$ are 'heavy' steel or brass particles. The material densities of each of these materials, alongside their *effective elasticities* can be seen in Table 1. The effective elasticity of a particle is determined through experimental measurements of the average fractional energy loss, due to *all sources*, of a particle during collisions with particles of the same species [12]. This binary system of spheres is housed within a fully three-dimensional cuboidal container and subjected to sinusoidal vibrations in the vertical direction, thus fluidising the granular bed. Unless otherwise specified, the bed is driven at a fixed frequency $f = 70$ Hz and amplitude $A = 1.06$ mm, giving a base velocity $V_0 = 0.47$ ms⁻². Square-based containers of width 25, 40, 50, 60, 80, 100, 120 and 160 mm are used, giving a range of aspect ratios $\in (0.04, 8)$. For all systems it is ensured that the height of the container is adequate to ensure the absence of particle collisions with the upper boundary, meaning that the system can be considered 'open'. For each experimental reading, it is ensured that the system is initially well mixed. Before data analysis begins, the system is excited for a period of 1000 s, allowing a steady state to be reached such that the equilibrium distribution of particles can be accurately measured. Each run is conducted over a period $t_{run} \gtrsim 3600$ s; runs corresponding to denser beds, within which the tracer will take longer to explore the system in its entirety, are conducted over considerably longer periods, in order to ensure good statistics. The steady state of a system is confirmed by splitting each run into a series of 50–500 s segments, the duration once again being determined by the system's density and hence mobility. The state of the system (specifically its density and temperature distributions) for each of these segments is then compared and, if there is no statistically significant fluctuation in either density or temperature, a steady state is assumed. The comparison of these segments also serves as a useful check of the reliability and reproducibility of the results obtained, as each of these individual temporal slices can be considered a data set in its own right. Nonetheless, for each set of system parameters, each full data set was also repeated in its entirety at a different point in time in order to account for possible differences in system behaviour due to subtle changes in conditions from day to day.

Data was acquired using positron emission particle tracking (PEPT), a non-invasive technique utilising a single tracer particle to analyse the time-averaged behaviour of the system. In order to perform PEPT, a single particle from the system is 'tagged' with a β^+ -emitting radioisotope. The emitted positrons rapidly annihilate within the dense medium of the tracer, producing pairs of back-to-back, 511 keV gamma rays. When the system is placed between the detector heads of a dual-headed gamma camera, these pairs of gamma rays can be used to triangulate the position of the tracer particle. With adequate activity, a succession of such location events can be used to record the three-dimensional motion of the tracer with millisecond time resolution and spatial resolution on the millimetre scale [13]. For steady state, ergodic systems, the time-averaged behaviour of the tracer particle can be used to determine various quantities such as particle velocity and mean squared displacement, as well as density and temperature profiles

[14]. For a binary system such as the one discussed here, two identical runs are performed for each system, one using a 'light' particle as the tracer and one using a 'heavy' particle, the combination of both data sets giving information corresponding to the entire system [15]. Further information regarding the PEPT technique and the manner in which the quantities relevant to this current study are calculated can be found in references [13,16] and [14,17–19] respectively.

It should be noted that, for all data presented, the absence of convective motion (which can strongly affect density-driven segregation within a system [20]) was ensured, thus allowing effects due to variations in aspect ratio to be isolated. In order to minimise the likelihood of the formation of convection rolls, 0.5 g of graphite powder was added to each system to act as a lubricant [21] and hence reduce friction. Since convective motion is, for dense and moderately dilute systems, heavily dependent on friction, the addition of such a lubricant results in a lower probability of convective motion occurring. Since convection has been previously shown to be additionally dependent on various system parameters, including the initial height of the bed, and the strength with which it is driven [22,23], the existence of convective motion was assessed for each individual data set. The absence of convective motion was confirmed in two manners: firstly, two-dimensional velocity vector fields for multiple 'slices' in the x - z and y - z planes were analysed. From these plots the presence of coordinated motion, such as that due to convection rolls, may easily be observed, as illustrated in Fig. 1. It is worth noting that, due to PEPT's use of highly penetrating γ -rays, particle motion is easily observable even deep within the bulk of a granular system. Secondly, the Fourier transform of the tracer particle's vertical motion is taken. The resulting spectrum can then be inspected to ensure the absence of any peaks corresponding to periodic motion within the system. From this Fourier analysis we are able to ensure not only the absence of convection but also the absence of any correlated motion between the vibrating base and the particles within the granular bed. Any data sets for which convective motion was found to be present were discarded.

2.2. Simulations

In addition to experimental results, data was also acquired from discrete particle simulations using the University of Twente's MercuryDPM code [24–27]. Wherever possible, experimental values of the relevant system parameters – including the particle numbers N_L and N_H , the frequency and amplitude, f and A , with which the system is excited and the particle size, d , density, ρ and effective elasticity for intra-species collisions, ϵ_{AA} – were implemented. The effective elasticity for inter-species collisions, ϵ_{AB} , was taken simply as the geometric average of the individual values for single species collisions, i.e. $\epsilon_{AB} = (\epsilon_{AA} + \epsilon_{BB})/2$. This definition of ϵ_{AB} , although simple, is a natural consequence of the spring-dashpot model of particle restitution [28,29]. The frictional coefficient μ is, unless stated otherwise, taken as 0.1 [30], and the elastic coefficient for particle collisions with the walls and base of the system is taken as $\epsilon_w = 0.6$.

3. Results and discussion

Due to the steady-state nature of the systems under investigation, the local packing fraction for a given region of the experimental system can be easily extrapolated from the single-particle data acquired using PEPT. By subdividing the container into a number of equally-sized spatial regions, the fractional residence time, F , of the tracer particle within each of these regions, or 'cells', can be determined.¹ Due to the

Table 1
Effective elasticities, $\epsilon_{\alpha\alpha}$, and material densities, ρ , for the various particles used in experiment.

Particle material	ρ (kg m ⁻³)	$\epsilon_{\alpha\alpha}$
Glass	2500	0.83
Aluminium	2700	0.69
Steel	7850	0.79
Brass	8500	0.61

¹ It is perhaps worth noting that, due to the highly penetrating nature of the 511 keV γ -rays with which PEPT is performed, particle motion can be tracked and recorded even deep within the interior of large, opaque and/or highly dense systems.

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